REVIEWS AND ANALYSES

Field Studies on Exposure, Effects, and Risk Mitigation of Aquatic Nonpoint-Source Insecticide Pollution: A Review

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ABSTRACT

Recently, much attention has been focused on insecticides as a group of chemicals combining high toxicity to invertebrates and fishes with low application rates, which complicates detection in the field. Assessment of these chemicals is greatly facilitated by the description and understanding of exposure, resulting biological effects, and risk mitigation strategies in natural surface waters under field conditions due to normal farming practice. More than 60 reports of insecticide-compound detection in surface waters due to agricultural nonpoint-source pollution have been published in the open literature during the past 20 years, about one-third of them having been undertaken in the past 3.5 years. Recent reports tend to concentrate on specific routes of pesticide entry, such as runoff, but there are very few studies on spray drift–borne contamination. Reported aqueous-phase insecticide concentrations are negatively correlated with the catchment size and all concentrations of >10 μg/L (19 out of 133) were found in smaller-scale catchments (<100 km^2). Field studies on effects of insecticide contamination often lack appropriate exposure characterization. About 15 of the 42 effect studies reviewed here revealed a clear relationship between quantified, non-experimental exposure and observed effects in situ, on abundance, drift, community structure, or dynamics. Azinphos-methyl, chlorpyrifos, and endosulfan were frequently detected at levels above those reported to reveal effects in the field; however, knowledge about effects of insecticides in the field is still sparse. Following a short overview of various risk mitigation or best management practices, constructed wetlands and vegetated ditches are described as a risk mitigation strategy that have only recently been established for agricultural insecticides. Although only 11 studies are available, the results in terms of pesticide retention and toxicity reduction are very promising. Based on the reviewed literature, recommendations are made for future research activities.

Agricultural pesticides are indispensable in modern farming. They are highly beneficial to the crops being grown, but their effects are less than desirable when they leave the target compartments of the agricultural ecosystem. Any unintended loss of pesticide is not only wasteful, but also represents a reduced efficiency and incurs increased costs to the user and the nontarget environment (Bowles and Webster, 1995; Falconer, 1998). Nonpoint-source pesticide pollution from agricultural areas is widely regarded as one of the greatest causes of contamination of surface waters (Gangbazo et al., 1999; Humenik et al., 1987; Line et al., 1997; Loague et al., 1998). Various routes of nonpoint-source pesticide transport into surface waters have been addressed (Baker, 1983; Edwards, 1973; Groenendijk et al., 1994; Schiavon et al., 1995).

Surface runoff due to rainfall events has attracted the most attention and several studies have summarized data on pesticides in runoff (Baker, 1983; Leonard, 1990; Wauchope, 1978; Willis and McDowell, 1982). Edge-of-field losses of pesticides range from less than 1% of the amount applied to 10% or more. Losses are greatest when severe rainstorms occur soon after pesticide application. The relative importance of sediment transport versus runoff water depends primarily on the soil adsorption properties of the pesticide (Wauchope, 1978). The potential for pesticide input into surface water following passage through the soil, including drainage transport, has been reviewed by Flury (1996). Particularly in loamy soils, there is evidence that even strongly adsorbed chemicals can move along preferential flow pathways. Although a direct comparison appears difficult, Flury (1996) concluded that the mass lost by leaching seems generally to be smaller than that lost by runoff, depending of course on the slope of the fields.

There are several generic scenarios for spray drift and spray deposition on surface waters. A large number of standardized drift studies conducted in Germany have been summarized by Ganzelmeier et al. (1995) and updated by Rautmann et al. (2001). The results were used to derive basic drift values widely used in European Union countries for regulatory risk assessment and 95th- or 90th-percentile values for deposited drift material for distances between 3 and 250 m. On the other hand, the Spray Drift Task Force's (SDTF) data set was analyzed and used to develop generic deposition curves with 95% confidence limits for distances between 0 and 549 m (USEPA, 1999a), which are proposed for use in risk assessment. Short- or long-range atmospheric transport with subsequent deposition into surface waters has recently been reported as a route of entry for current-use pesticides into the Sierra Nevada (Le Noir et al., 1999), but not enough information is available to assess its importance. In addition to measurement of actual exposure concentrations, models that predict exposure to pesticides in surface waters have been developed and are currently used in ecological risk assessment based on worst-case and probabilistic scenarios (Adriaanse et al., 1997; Groenendijk et al., 1994; Hart, 2001).

Among the various types of pesticides that potentially

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contaminate surface water, insecticides play an important role in aquatic ecosystems as documented by the accumulated data on their detrimental effects to community structure, reproduction, and developmental processes among several taxa including macroinvertebrates, amphibians, birds, fish, and other wildlife (Colborn et al., 1993; Scott et al., 1987; Thompson, 1996). According to the database of the United States National Center for Food and Agricultural Policy, the use of insecticides in the USA has increased by 18.2%, from 67 116 metric tons in 1992 to 82 080 metric tons of active ingredient in 1997 (National Center for Food and Agricultural Policy, 1997). Due to their relatively high toxicity to aquatic fauna (Brock et al., 2000), many insecticides are regarded as priority pollutants among the variety of chemicals entering aquatic systems via nonpoint sources. From a review of a pesticide risk reduction program in Ontario, Canada using data collected from 1973 to 1998, Gallivan et al. (2001) concluded that major reductions in risk can be achieved by reducing the use of high-risk pesticides (e.g., insecticides) on fruit and vegetables.

As for most pesticides, there are numerous reports related to the single-species laboratory toxicity of insecticides (USEPA, 1995). The microcosm and mesocosm studies available have recently been summarized and reviewed by Brock et al. (2000). Keeping in mind that the ultimate scientific goal in the ecological risk assessment of pesticides is to understand and assess potential effects under field conditions, there is a need for exposure and effect studies conducted in natural surface waters affected by normal farming practices. From a limnological point of view, Schindler (1998) has compared the results of bottle and mesocosm experiments with whole-ecosystem experiments using the Experimental Lake Area (ELA) in northwestern Ontario, Canada. He concluded that the upscaling from mesocosm to whole lakes and even from small lakes to bigger ones may result in considerable shortcomings and misinterpretations due to major differences in spatial and temporal scales. Moreover, biochemical and fitness differences in sensitivity to insecticides of field and laboratory-derived populations of midge (*Chironomus riparius*) have been reported (Hoffman and Fisher, 1994), further illustrating the difficulties in the translation of experimental results to natural environments.

Although laboratory tests using aquatic organisms are of unquestionable benefit in assessing the hazard of pesticides to aquatic ecosystems, the simplistic environmental conditions under which they are often conducted limit their predictive capability. In the early 1980s, Koeman (1982) emphasized developing test systems that reflect a greater complexity. Although multispecies approaches, subsequently developed, eliminated some of these problems, these protocols still suffer from inherent limitations when laboratory results are extrapolated to predicted effects on natural aquatic ecosystems. Ecosystems are typically affected by several stressors (e.g., varying water levels, habitat alterations, chemical pollution) simultaneously, and the intensity of each varies through space and time. Cumulative effects of these multiple effects are altered by synergistic and antagonistic interactions among individual stressors and between anthropogenic and natural perturbations. Thus, it is essential that predictions derived from experimental approaches be validated in natural ecosystems and that long-term monitoring efforts be implemented to ensure that unexpected long-term ecosystem effects do not occur (Cairns et al., 1994). Ecosystem-level information is not only relevant to the effects of pollutants, but is also considered beneficial for exposure assessment in facilitating the monitoring of pollutant presence in environmental compartments (Touart and Maciorowski, 1997; Van Dijk et al., 2000).

Although some reviews or summary reports on the presence of insecticides in various nonpoint routes have been published (e.g., Ganzelmeier et al., 1995; Wauchope, 1978), there are almost no such studies addressing work that has been done on the presence or effects of insecticides in the receiving surface waters. Willis and McDowell (1982) listed toxicity data and physicochemical properties for pesticides that occur in surface runoff. An overview of the biological effects of agriculturally derived surface-water pollutants is given by Cooper (1993). Conservation tillage in relation to pesticide runoff in surface waters is generally summarized by Fawcett et al. (1994) and what is known about the ecotoxicology of wetlands has recently been summarized (Lewis et al., 1999). Studies on pesticide ecotoxicology in tropical aquatic habitats in Central America were summarized by Castillo et al. (1997), with emphasis on the pesticide contents in the biota, and Clark et al. (1993) reported ecotoxicological examples from coastal wetlands. From all these reviews, the lack of data referring to insecticide exposure, effects, and risk mitigation under field conditions is apparent.

AIMS

The purpose of this review is to:

- compile and interpret the results of case studies in which nonpoint-source insecticide contamination, resulting from normal farming practices, was measured in aquatic habitats, to describe the exposure situation;
- compile and interpret the results of field studies in which the effects of insecticides were measured in aquatic habitats under normal agricultural practice, to describe the effect situation;
- evaluate the field situation through a comparison of exposure and effect studies;
- compile and interpret the results of field studies on the use of constructed wetlands as a risk mitigation strategy for aquatic nonpoint-source insecticide pollution; and
- propose directions for future research efforts.

EXPOSURE

There are various studies reporting the pesticide contamination of surface waters on a national or even international scale. Results of the United States National Water Quality Assessment (NAWQA) Program performed by the U.S. Geological Survey were reported by Larson et al. (1999). The data available for northern European countries such as Norway, Sweden, and Finland have been summarized on behalf of the European Union by Lundbergh et al. (1995), those for the Netherlands by the Committee for Integral Water Management/Committee for the Enforcement of the Water Pollution Act (1995) and Teunissen-Ordelmann and Schrap (1997), those for England and Wales by Environment Agency (2000), and those for Germany by the Federal Environmental Agency (Federal Environmental Agency, 1999) and Zullei-Seibert (1990). Some data for Denmark are reviewed by Mogensen and Spliid (1995), but this report deals exclusively with herbicides. However, most of these studies are based on regular governmental monitoring programs and are not discussed further in this review.

Table 1 lists case studies published since 1982 on the detection of insecticides in surface waters due to agricultural nonpoint-source pollution. The reports are sorted according to the insecticide compound; for a given compound detections in water are listed first, followed by detections in suspended particles and sediments. There are numerous studies published before 1982 that are not included in Table 1, most of them dealing with organochlorine insecticides (e.g., Bradley et al., 1972; Cope, 1966; Croll, 1969; Gorbach et al., 1971; Greichus et al., 1977; Greve, 1972; Heckman, 1981; Herzel, 1971; Jackson et al., 1974; Kuhr et al., 1974; Miles, 1976; Miles and Harris, 1971, 1973; Pollero et al., 1976; Richard et al., 1975). Ramesh et al. (1991) gave a short overview of exemplary studies on organochlorine contamination in surface waters. In 1960, a study on the input of parathion into a farm pond in South Carolina was started (Nicholson et al., 1962), which is regarded as one of the pioneer investigations on insecticides in agricultural surface waters.

During the past two decades, the number of published studies has increased continuously. A total of 10 studies were reported in the 7-yr period between 1982 and 1989, while 15 and 24 studies were published in the subsequent 5-yr periods between 1990 and 1994 and between 1995 and 1999. Finally, a total of 23 studies came out in the period of only 3.5 yr between 2000 and the middle of 2003. The number of times an insecticide was detected also increased and was 33, 37, 56, and 58 in the respective time periods, while the number of insecticide compounds that were detected did not show any particular trend, with 21, 16, 24, and 16 different compounds, respectively. Although these numbers refer only to studies available in the open literature, they demonstrate a clear increase of scientific interest in the topic, probably driven by the development of modern analytical methods, such as gas chromatography–mass spectrometry, gas chromatography–tandem mass spectrometry, or liquid chromatography–tandem mass spectrometry and by the increasing need for data on pesticide exposure to assess human and environmental health.

On the other hand, the detection of low levels of pesticides in river water is considerably more difficult than in many other types of waters. This is illustrated by results from a large monitoring data set compiled by German drinking-water authorities (Zullei-Seibert, 1990). In relatively easy-to-analyze matrices, such as drinking water, ground water, dam water, and spring water, pesticides were detected about five times more often below the European drinking water threshold level for individual pesticide compounds of 0.1 μg/L than above this level, which can be simply explained by the higher likelihood of low-level pollution occurring and thus being detected. Although similar results would be expected, for river water twice as many detections were above the 0.1 μg/L level than below, suggesting a non-negligible matrix influence of the type of water to be analyzed. It follows from these results that many low-level contaminations are presumably not detected in agricultural rivers and lakes, simply because of the matrix influence. A further aspect adding to the difficulty to detect current-use insecticide contamination is the fact that the toxicity of modern insecticides, such as pyrethroids, is generally higher than for older groups of compounds. Therefore, lower application rates are used to obtain the same level of pest control, resulting also in lower-level contamination in the environment, which is considerably more difficult to detect.

Results of an extensive program on pesticide loss to stream water from agricultural areas of the Great Lakes catchment in Ontario, Canada revealed the presence of carbofuran, chlorpyrifos, diazinon, endosulfan, and ethion (Table 1) in water samples at levels up to 3.8 μg/L (Frank et al., 1982; Richards and Baker, 1993). Later investigations focused on the contamination of farm wells with pesticides (Frank et al., 1990, 1987a, 1987b). Various studies in British Columbia, Canada were stimulated by detection of endosulfan at a very high concentration of 1530 μg/L in ditch water during spray application on adjacent fields. Levels in sediments varied in affected ditches between 2 and 150 μg/kg, with an average of 18.8 μg/kg (Wan, 1989). A follow-up study focused on organophosphate insecticides, of which diazinon, dimethoate, fensulfothion, and parathion (Table 1) were detected in farm ditches channeling the discharge from vegetable and field crop areas (Wan et al., 1994). Later, Wan et al. (1995a; Table 1) reported on extensive data on concentrations of endosulfan in soils, ditch water, and sediments (Wan et al., 1995a; Table 1) as well as azinphos-methyl and parathion-ethyl losses from cranberry (*Vaccinium oxycoccos* L.) bogs (Wan et al., 1995b), which led to peak levels of 175 and 21 μg/L, respectively, in the adjacent surface water.

Cooper and coworkers at the USDA Agricultural Research Service's National Sedimentation Laboratory in the 1970s initiated studies on the effects of agricultural erosion on aquatic ecosystems in the lower Mississippi River catchment (Cooper, 1987; Cooper and Bacon, 1980; Cooper and Knight, 1986; Cooper et al., 1993; Dendy, 1983), which they later extended to the detection of pesticide contamination in various surface water ecosystems. Residual concentrations of insecticides such as DDT and toxaphene were reported from fishes, surface water, and sediments (Cooper et al., 1987; Cooper and Knight, 1987). Other studies took place in the Moon Lake, a 10.1-km^2 oxbow lake of the Mississippi River, and measured the current-use insecticides fenvalerate (0.11 μg/L and 10.8 μg/kg), permethrin (0.13 μg/L), and

Table 1. Summary of field case studies on insecticide contamination in surface waters due to agricultural practice published since 1982.

Substance	Chemical name	Concentration†	Source	Number of detections	Sampling interval	Location	Catchment size	Reference
							km²	
Aldicarb	2-methyl-2-(methylthio)propionaldehyde *O*-methylcarbamoyloxime	0.5–6.4 μg/L	nonpoint sources	92	monthly	Upper Jordan Basin, Israel	300	Bar-Ilan et al. (2000)
Aldicarb		0.21–2.84 μg/L	runoff	2	event	ADAS Rosemaund, UK	approx. 0.35	Williams et al. (1995)
Azinphos-methyl	*S*-(3,4-dihydro-4-oxobenzo[*d*]-[1,2,3]-triazin-3-ylmethyl) *O*,*O*-dimethyl phosphorodithioate	0.001–0.2 μg/L	leaching (irrigation)	12	weekly	Royal Lake, Washington	6 400	Gruber and Munn (1998)
Azinphos-methyl		0.001–0.016 μg/L	nonpoint sources	20	60 d	Ioannina Lake, Greece	1 330	Albanis et al. (1986)
Azinphos-methyl		0.001–0.025 μg/L	nonpoint sources	12	60 d	Kalamas River, Greece	1 330	Albanis et al. (1986)
Azinphos-methyl		0.06–1.0 μg/L	nonpoint sources	7	single	orchard wetlands, Ontario, Canada	approx. 5	Harris et al. (1998)
Azinphos-methyl		0.02–0.1 μg/L	runoff	3	event	Berg and Franschoek Rivers, South Africa	20–150	Schulz (2003)
Azinphos-methyl		0.07–0.38 μg/L	runoff	3	event	Lourens River, South Africa	92	Schulz et al. (2001a)
Azinphos-methyl		0.06–1.5 μg/L	runoff	5	event	Lourens River and tributaries, South Africa	92	Schulz (2001b)
Azinphos-methyl		0.39–0.6 μg/L	runoff	5	event	six Lourens River subcatchments, South Africa	0.15–1	Dabrowski et al. (2002a)
Azinphos-methyl		0.14–0.8 μg/L	runoff	4	event	Lourens River tributary, South Africa	0.15	Schulz and Peall (2001)
Azinphos-methyl		0.1–7 μg/L	runoff	11	event	three estuarine sites, South Carolina	10–30	Scott et al. (1999)
Azinphos-methyl		0.002–21 μg/L	runoff	–	event	estuarine sites, South Carolina	10–100	Finley et al. (1999)
Azinphos-methyl		3.4–244.6 μg/kg SP	runoff	5	event	Lourens River, South Africa	92	Schulz et al. (2001a)
Azinphos-methyl		216–1247 μg/kg SP	runoff	2	event	Lourens River and tributaries, South Africa	92	Schulz (2001b)
Azinphos-methyl		21.1 μg/kg SP	runoff	1	event	six Lourens River subcatchments, South Africa	0.15–1	Dabrowski et al. (2002a)
Azinphos-methyl		0.9–43.3 μg/kg SP	runoff	2	14 d	Lourens River tributary, South Africa	0.15	Schulz and Peall (2001)
Azinphos-methyl		1.1–2.6 μg/L	spray drift	6	event	Lourens River tributary, South Africa	0.15	Schulz et al. (2001b)
Azinphos-methyl		0.03–0.05 μg/L	spray drift	3	event, composite	Lourens River, South Africa	4	Schulz et al. (2001b)
Azinphos-methyl		0.36–0.87 μg/L	spray drift	5	event	Lourens River tributary, South Africa	0.15	Schulz et al. (2001c)
Carbaryl	1-naphthyl methylcarbamate	0.07–0.14 μg/L	leaching, runoff	2	monthly	seven sites, Pajaro River estuary, California	approx. 140	Hunt et al. (1999)
Carbaryl		0.1–0.59 μg/L	application to rice fields	approx. 6	weekly	Shinano River, Japan	11 900	Tanabe et al. (2001)
Carabryl		0.003–0.2 μg/L	runoff	–	event	streams in the U.S. Midwest	200–443 670	Battaglin and Fairchild (2002)
Carbofuran	2,3-dihydro-2,2-dimethylbenzofuran-7-yl methylcarbamate	0.001–0.042 μg/L	nonpoint sources	22	60 d	Ioannina Lake, Greece	1 330	Albanis et al. (1986)
Carbofuran		0.001–0.012 μg/L	nonpoint sources	12	60 d	Kalamas River, Greece	1 330	Albanis et al. (1986)
Carbofuran		4.8 μg/L	nonpoint sources	1	monthly	Sacramento–San Joaquin catchment, California	40 000	Werner et al. (2000)

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Table 1. Continued.

Substance	Chemical name	Concentration†	Source	Number of detections	Sampling interval	Location	Catchment size	Reference
							km²	
Carbofuran		0.1–1.8 μg/L	nonpoint sources	8	weekly, composite	11 agricultural watersheds, Ontario, Canada	40	Frank et al. (1982)
Carbofuran		0.02–49.4 μg/L	runoff	4	event	ADAS Rosemaund, UK	approx. 0.35	Williams et al. (1995)
Carbofuran		0.003–1.03 μg/L	runoff	–	event	streams in the U.S. Midwest	200–443 670	Battaglin and Fairchild (2002)
Chlorpyrifos	*O,O*-diethyl *O*-3,5,6-trichloro-2-pyridyl phosphorothioate	0.004–0.12 μg/L	leaching (irrigation)	15	weekly	Royal Lake, Washington	6 400	Gruber and Munn (1998)
Chlorpyrifos		0.06–0.52 μg/L	nonpoint sources	8	monthly	Sacramento–San Joaquin catchment, California	approx. 40 000	Werner et al. (2000)
Chlorpyrifos		0.01–1.6 μg/L	nonpoint sources	6	weekly, composite	11 agricultural watersheds, Ontario, Canada	40	Frank et al. (1982)
Chlorpyrifos		0.004–0.86 μg/L	runoff	–	event	streams in the U.S. Midwest	200–443 670	Battaglin and Fairchild (2002)
Chlorpyrifos		0.13 μg/L	runoff	1	14 d	White River, Indiana	29 383	Chen et al. (2002)
Chlorpyrifos		0.03–0.2 μg/L	runoff	2	event	Lourens River and tributaries, South Africa	92	Schulz (2001b)
Chlorpyrifos		0.01–0.26 μg/L	runoff	17	event	San Joaquin catchment, California	approx. 16 000	Domagalski et al. (1997)
Chlorpyrifos		0.19 μg/L	runoff	1	event	six Lourens River subcatchments, South Africa	0.15–1	Dabrowski et al. (2002a)
Chlorpyrifos		0.01–0.03 μg/L	runoff	2	event	Lourens River tributary, South Africa	0.15	Schulz and Peall (2001)
Chlorpyrifos		0.08–1.3 μg/L	runoff	3	event	Lourens River tributary, South Africa	0.15	Moore et al. (2002)
Chlorpyrifos		0.03–3.2 μg/L	runoff	52	event	creek channel, California	approx. 150	Hunt et al. (2003)
Chlorpyrifos		0.02–3.8 μg/L	runoff	7	8 h	Sandusky River, OH	3 240	Richards and Baker (1993)
Chlorpyrifos		1.0–2.1 μg/kg SP	runoff	7	24–48 h, event	San Joaquin–Sacramento estuary, California	approx. 40 000	Bergamaschi et al. (2001)
Chlorpyrifos		2–344.2 μg/kg SP	runoff	8	event	Lourens River, South Africa	92	Schulz et al. (2001a)
Chlorpyrifos		83–924 μg/kg SP	runoff	2	event	Lourens River and tributaries, South Africa	92	Schulz (2001b)
Chlorpyrifos		4.2–152 μg/kg SP	runoff	8	event	six Lourens River subcatchments, South Africa	0.15–1	Dabrowski et al. (2002a)
Chlorpyrifos		0.2–31.4 μg/kg SP	runoff	2	14 d	Lourens River tributary, South Africa	0.15	Schulz and Peall (2001)
Chlorpyrifos		69–720 μg/kg SP	runoff	6	event	Berg and Franschoek Rivers, South Africa	20–150	Schulz (2003)
Chlorpyrifos		2.6–89.4 μg/kg SP	runoff	3	event	Lourens River tributary, South Africa	0.15	Moore et al. (2002)
Chlorpyrifos		0.2–2.8 μg/L	runoff, assumed	7	1 d (peak)	Sandusky River, Ohio	3 200	Giesy et al. (1999)
Chlorpyrifos		0.67 μg/L	runoff, assumed	7	1 d (peak)	Turlock Irrigation Ditch, California	approx. 30	Giesy et al. (1999)
Chlorpyrifos		0.05–0.1 μg/L	runoff, impregnated bags	9	seasonal	Suerte River tributaries, Costa Rica	57	Castillo et al. (2000)
Cyfluthrin	(*RS*)-α-cyano-4-fluoro-3-phenoxybenzyl (1 *RS*,3*RS*;1*RS*,3*SR*)-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylate	0.2–5 μg/L	nonpoint sources	7	daily	Vemmenhög subcatchment, southern Sweden	8.3	Kreuger (1998)
Cypermethrin	(*RS*)-α-cyano-3-phenoxybenzyl (1 *RS*,3*RS*; 1*RS*,3*SR*)-3-(2,2-dichlorovinyl)-2,2-dimethylcyclopropanecarboxylate	0.4–1.7 μg/L	spray drift	20	event	experimental vineyards, France	approx. 0.1	Crossland et al. (1982)
Cypermethrin		2.7 μg/kg	nonpoint sources	1	single	five streams and ditches, southern UK	approx. 10	House et al. (1991)

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Table 1. Continued.

Substance	Chemical name	Concentration†	Source	Number of detections	Sampling interval	Location	Catchment size	Reference
							km²	
DDT	1,1,1-trichloro-2,2-bis(4-chlorophenyl)ethane	0.13–1.1 μg/L	leaching, runoff	7	monthly	seven sites, Pajaro River estuary, California	approx. 140	Hunt et al. (1999)
DDT		0.01–0.6 μg/L	nonpoint sources	124	monthly	five sites, Dear Creek, Mississippi	44	Cooper et al. (1987)
DDT		1.1–4.7 μg/kg SP	nonpoint sources	2	single	River Windrush catchment, southern UK	approx. 150	House et al. (1992)
DDT		1–8.5 μg/kg	nonpoint sources	8	seasonal	four sites, Vellar River, southern India	3 200	Ramesh et al. (1991)
DDT		0.6–62.2 μg/kg	nonpoint sources	5	single	five streams and ditches, southern UK	approx. 10	House et al. (1991)
DDT		0.2 μg/kg	nonpoint sources	1	single	River Windrush catchment, southern UK	approx. 150	House et al. (1992)
DDT		0.01–650.8 μg/kg	runoff	45	single	Moon Lake catchment, Mississippi	166	Cooper (1991b)
Deltamethrin	(*S*)-α-cyano-3-phenoxybenzyl (1*R*,3*R*)-3-(2,2-dibromovinyl)-2,2-dimethyl-cyclopropanecarboxylate	1.9–37.5 μg/kg	nonpoint sources	3	single	five streams and ditches, southern UK	approx. 10	House et al. (1991)
Deltamethrin		0.08–2 μg/L	runoff	3	event	ADAS Rosemaund, UK	0.34	Turnbull et al. (1995)
Deltamethrin		1.4 μg/L	runoff	1	event	six Lourens River subcatchments, South Africa	0.15–1	Dabrowski et al. (2002a)
Diazinon	*O*,*O*-diethyl *O*-2-isopropyl-6-methylpyrimidin-4-yl phosphorothioate	0.1–0.32 μg/L	application to rice fields	approx. 15	weekly	Shinano River, Japan	11 900	Tanabe et al. (2001)
Diazinon		0.05–1.06 μg/L	leaching, runoff	7	monthly	seven sites, Pajaro River estuary, California	approx. 140	Hunt et al. (1999)
Diazinon		0.001–0.057 μg/L	nonpoint sources	26	60 d	Ioannina Lake, Greece	1 330	Albanis et al. (1986)
Diazinon		0.002–0.052 μg/L	nonpoint sources	12	60 d	Kalamas River, Greece	1 330	Albanis et al. (1986)
Diazinon		0.4 μg/L	nonpoint sources	1	monthly	Sacramento–San Joaquin catchment, California	approx. 40 000	Werner et al. (2000)
Diazinon		0.1–0.3 μg/L	nonpoint sources	3	seasonal	Edo River, Japan	200	Kikuchi et al. (1999)
Diazinon		0.01–0.51 μg/L	nonpoint sources	50	monthly	six farm ditches, British Columbia, Canada	approx. 0.5	Wan et al. (1994)
Diazinon		0.15 μg/L	nonpoint sources	1	weekly, composite	11 agricultural watersheds, Ontario, Canada	40	Frank et al. (1982)
Diazinon		0.05–0.4 μg/L	nonpoint sources	5	seasonal	San Joaquin–Sacramento estuary, California	approx. 40 000	Domagalski and Kuivila (1993)
Diazinon		0.02–0.62 μg/L	nonpoint sources	7	weekly	San Joaquin catchment, California	approx. 16 000	Domagalski et al. (1997)
Diazinon		0.12–7 μg/L	runoff	17	event	San Joaquin tributaries, California	50–100	Domagalski et al. (1997)
Diazinon		0.02–1.03 μg/L	runoff	approx. 60	daily	Sacramento–San Joaquin catchment, California	approx. 40 000	Kuivila and Foe (1995)
Diazinon		0.07–0.15 μg/L	runoff, impregnated bags	7	seasonal	Suerte River tributaries, Costa Rica	57	Castillo et al. (2000)
Diazinon		0.1–1.5 μg/kg SP	nonpoint sources	5	seasonal	San Joaquin–Sacramento estuary, California	approx. 40 000	Domagalski and Kuivila (1993)
Dichlorvos	2,2-dichlorovinyl dimethyl phosphate	0.1–0.3 μg/L	nonpoint sources	8	seasonal	Tama River, Japan	1 240	Kikuchi et al. (1999)
Dicofol	2,2,2-trichloro-1,1-bis(4-chlorophenyl)ethanol	0.2–2.5 μg/L	leaching (irrigation)	6	bimonthly	Orestimba Creek, California	approx. 200	Domagalski (1996)
Dieldrin	(1*R*,4*S*,4a*S*,5*R*,6*R*,7*S*,8*S*,8a*R*)-1,2,3,4,10,10-hexachloro-1,4,4a,5,6,7,8,8a-octahydro-6,7-epoxy-1,4:5,8-dimethanonaphthalene	0.06–0.26 μg/L	leaching, runoff	4	monthly	seven sites, Pajaro River estuary, California	approx. 140	Hunt et al. (1999)
Dieldrin		8–17 μg/kg SP	nonpoint sources	3	single	River Windrush catchment, southern UK	approx. 150	House et al. (1992)
Dieldrin		1.0–6.7 μg/kg	nonpoint sources	4	single	five streams and ditches, southern UK	approx. 10	House et al. (1991)

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Table 1. Continued.

Substance	Chemical name	Concentration†	Source	Number of detections	Sampling interval	Location	Catchment size	Reference
							km²	
Dieldrin		0.2 μg/kg	nonpoint sources	2	single	River Windrush catchment, southern UK	approx. 150	House et al. (1992)
Dimethoate	*O,O*-dimethyl *S*-methylcarbamoylmethyl phosphorodithioate	0.2 μg/L	application to rice fields	4	weekly	Shinano River, Japan	11 900	Tanabe et al. (2001)
Dimethoate		0.05–0.1 μg/L	nonpoint sources	2	single	San Joaquin River and tributaries, California	100–5 000	Pereira et al. (1996)
Dimethoate		0.1–30 μg/L	nonpoint sources	7	daily	Vemmenhög subcatchment, southern Sweden	8.3	Kreuger (1998)
Dimethoate		0.01–11.6 μg/L	nonpoint sources	34	monthly	six farm ditches, British Columbia, Canada	approx. 0.5	Wan et al. (1994)
Disulfoton	*O,O*-diethyl *S*-2-ethylthioethyl phosphorodithioate	0.1–0.4 μg/L	runoff	8	event	Shell Creek, Nebraska	700	Spalding and Snow (1989)
Endosulfan	6,7,8,9,10,10-hexachloro-1,5,5a,6,9,9a-hexahydro-6,9-methano-2,4,3-benzodioxathiepine 3-oxide	0.05–0.53 μg/L	nonpoint sources	3	single	orchard wetlands, Ontario, Canada	approx. 5	Harris et al. (1998)
Endosulfan		0.06–0.75 μg/L	nonpoint sources	18	monthly	27 agricultural channel sites, southern Florida	400	Miles and Pfeuffer (1997)
Endosulfan		0.01–0.17 μg/L	nonpoint sources	365	weekly, composite	11 agricultural watersheds, Ontario, Canada	40	Frank et al. (1982)
Endosulfan		0.1 μg/L (maximum)	nonpoint sources	1	monthly	up to 29 streams, southern Sweden	2–500	Kreuger and Brink (1988)
Endosulfan		0.01–13.4 μg/L	nonpoint sources	61	periodic	seven sites, farm ditches, British Columbia, Canada	1–12	Wan et al. (1995a)
Endosulfan		1 530 μg/L	spray drift	1	event	seven farm ditches, British Columbia, Canada	approx. 10	Wan (1989)
Endosulfan		0.03–0.16 μg/L	runoff	7	event	Lourens River, South Africa	92	Schulz et al. (2001a)
Endosulfan		0.06–2.9 μg/L	runoff	5	event	Lourens River and tributaries, South Africa	92	Schulz (2001b)
Endosulfan		4 μg/L	runoff	1	event	Gwydir River, southeastern Australia	approx. 10 000	Muschal (1998)
Endosulfan		0.11–2.0 μg/L	runoff	4	event	211 rural ponds, Ontario, Canada	10–80	Frank et al. (1990)
Endosulfan		0.01–0.85 μg/L	runoff	35	event	three estuarine sites, South Carolina	10–30	Scott et al. (1999)
Endosulfan		1.44 μg/L	runoff	1	event	Adams Creek, South Carolina	approx. 15	Ross et al. (1996)
Endosulfan		0.06–0.32 μg/L	runoff	10	event	six Lourens River subcatchments, South Africa	0.15–1	Dabrowski et al. (2002a)
Endosulfan		0.07–0.2 μg/L	runoff	2	event	Lourens River tributary, South Africa	0.15	Schulz and Peall (2001)
Endosulfan		17.7–24.6 μg/kg SP	runoff	2	24–48 h, event	San Joaquin–Sacramento estuary, California	approx. 40 000	Bergamaschi et al. (2001)
Endosulfan		3.9–245.3 μg/kg SP	runoff	6	event	Lourens River, South Africa	92	Schulz et al. (2001a)
Endosulfan		179–12 082 μg/kg SP	runoff	2	event	Lourens River and tributaries, South Africa	92	Schulz (2001b)
Endosulfan		9.7–273 μg/kg SP	runoff	8	event	six Lourens River subcatchments, South Africa	0.15–1	Dabrowski et al. (2002a)
Endosulfan		4.6–156 μg/kg SP	runoff	6	event	Berg and Franschoek Rivers, South Africa	20–150	Schulz (2003)
Endosulfan		10–318 μg/kg SP	nonpoint sources	4	event	two rural rivers, Argentina	50–100	Jergentz et al. (2004)
Endosulfan		334–926 μg/kg	nonpoint sources	5	periodic	seven farm ditch sites, British Columbia, Canada	approx. 10	Wan (1989)
Endosulfan		5–2 461 μg/kg	runoff	47	periodic	seven sites, farm ditches, British Columbia, Canada	1–12	Wan et al. (1995a)
Endosulfan		3–48 μg/kg	runoff	3	event	Namoi River and tributary, southeastern Australia	approx. 5 000	Leonard et al. (2001)
Endosulfan		8.5–12.3 μg/L	spray drift	3	event	Lourens River tributary, South Africa	approx. 0.5	Schulz et al. (2001b)

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Table 1. Continued.

Substance	Chemical name	Concentration†	Source	Number of detections	Sampling interval	Location	Catchment size	Reference
							km²	
Endosulfan		0.04–0.1 μg/L	spray drift	3	event, composite	Lourens River, South Africa	4	Schulz et al. (2001b)
Endosulfan α and β		1.5–69.4 μg/kg	nonpoint sources	3	single	orchard wetlands, Ontario, Canada	approx. 5	Harris et al. (1998)
Ethion	*O,O,O′,O′*-tetraethyl *S,S′*-methylene bis(phosphorodithioate)	0.01–0.04 μg/L	nonpoint sources	4	weekly, composite	11 agricultural watersheds, Ontario, Canada	40	Frank et al. (1982)
Fenitrothion	*O,O*-dimethyl *O*-4-nitro-*m*-tolyl phosphorothioate	0.2–0.4 μg/L	aerial application	7	14 d	River Onogawa, Japan	150	Takamura (1996)
Fenitrothion		0.1–1.7 μg/L	application to rice fields	approx. 22	weekly	Shinano River, Japan	11 900	Tanabe et al. (2001)
Fenitrothion		0.1–80 μg/L	application to rice fields	5	daily, weekly	Ono River, Japan	approx. 80	Hatakeyama et al. (1991)
Fenitrothion		0.1–0.2 μg/L	nonpoint sources	8	seasonal	Tama River, Japan	1 240	Kikuchi et al. (1999)
Fenitrothion		0.1 μg/L (maximum)	nonpoint sources	2	monthly	up to 29 streams, southern Sweden	2–500	Kreuger and Brink (1988)
Fenobucarb	2-*sec*-butylphenyl methylcarbamate	0.2–1.5 μg/L	aerial application	11	14 d	River Onogawa, Japan	150	Takamura (1996)
Fenobucarb		3.9–22.4 μg/L	application in rice fields	–	event	Kajinashi River, Japan	80	Tada and Shiraishi (1994)
Fenobucarb		0.1–1.3 μg/L	application to rice fields	approx. 30	weekly	Shinano River, Japan	11 900	Tanabe et al. (2001)
Fenobucarb		15.6–36.1 μg/L	application to rice fields	3	seasonal	subcatchment of River Koise, Japan	4–12	Iwakuma et al. (1993)
Fenobucarb		0.1–4 μg/L	application to rice fields	8	14 d	Sawato River, Japan	6	Takamura et al. (1991b)
Fensulfothion	*O,O*-diethyl *O*-4-methylsulfinylphenyl phosphorothioate	10.3 μg/kg	nonpoint sources	2	periodic	seven farm ditch sites, British Columbia, Canada	approx. 10	Wan (1989)
Fenthion	*O,O*-dimethyl-*O*-4-methylthio-*m*-tolyl phosphorothioate	0.5–50 μg/L	aerial application	12	event	Suna River, Japan	80	Hatakeyama and Yokoyama (1997)
Fenthion		0.2 μg/L	aerial application	8	14 d	River Onogawa, Japan	150	Takamura (1996)
Fenthion		0.05–65 μg/L	application to rice fields	13	daily, weekly	Bizen River, Japan	approx. 80	Hatakeyama et al. (1991)
Fenvalerate	(*RS*)-α-cyano-3-phenoxybenzyl (*RS*)-2-(4-chlorophenyl)-3-methylbutyrate	0.2–6.2 μg/L	runoff	7	event	Ohebach, northern Germany	1	Liess et al. (1999)
Fenvalerate		0.01–0.11 μg/L	runoff	9	biweekly	Moon Lake catchment, Mississippi	166	Cooper (1991b)
Fenvalerate		0.11 μg/L	runoff	1	event	Leadenwah Creek, South Carolina	approx. 2 000	Baughman et al. (1989)
Fenvalerate		0.02–0.9 μg/L	runoff	18	event	three estuarine sites, South Carolina	10–30	Scott et al. (1999)
Fenvalerate		302 μg/kg	runoff	1	event	Ohebach, northern Germany	1	Liess et al. (1999)
Fenvalerate		20–70 μg/kg SP	nonpoint sources	3	seasonal	Vemmenhög catchment, southern Sweden	9	Kreuger et al. (1999)
Fenvalerate		10–80 μg/kg	nonpoint sources	3	seasonal	Vemmenhög catchment, southern Sweden	9	Kreuger et al. (1999)
Fenvalerate		0.6–3.6 μg/kg	nonpoint sources	5	single	five streams and ditches, southern UK	approx. 10	House et al. (1991)
Fenvalerate		0.7–10.8 μg/kg	nonpoint sources	5	single	Moon Lake catchment, Mississippi	166	Cooper (1991a)
Fenvalerate		33–71.3 μg/kg SP	runoff	2	14 d	Ohebach, northern Germany	1	Liess et al. (1996)
Fenvalerate		1.0–10 μg/kg	runoff	3	event	Ohebach, northern Germany	1	Liess et al. (1999)
Fipronil	(±)-5-amino-1-(2,6-dichloro-α,α,α-(trifluoro-*p*-tolyl)-4-trifluoromethyl-sulfinylpyrazole-3-carbonitrile	9.1 μg/L	rice seed coating	1	single	Rice farms, Mississippi	approx. 50	Schlenk et al. (2001)

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Table 1. Continued.

Substance	Chemical name	Concentration†	Source	Number of detections	Sampling interval	Location	Catchment size	Reference
							km²	
Fipronil		5.5 μg/kg	rice seed coating	1	single	Rice farms, Mississippi	approx. 50	Schlenk et al. (2001)
Fonofos	*O*-ethyl *S*-phenyl (*RS*)-ethylphosphonodithioate	0.32 μg/L	runoff	1	14 d	White River, Indiana	29 383	Chen et al. (2002)
Fonofos		0.01–11.9 μg/L	runoff	89	8 h	Lost Creek, OH	11.3	Richards and Baker (1993)
Lindane	1,2,3,4,5,6-hexachlorocyclohexane (mixed isomers)	0.1–4.9 μg/L	nonpoint sources	4	monthly	Reconquista River, Argentina	1 670	Rovedatti et al. (2001)
Lindane		0.1 μg/L (average)	nonpoint sources	104	seasonal	Keoladeo wetland, India	approx. 30	Muralidharan (2000)
Lindane		0.6 μg/L (maximum)	nonpoint sources	8	monthly	up to 29 streams, southern Sweden	2–500	Kreuger and Brink (1988)
Lindane		0.03–0.8 μg/L	nonpoint sources	7	monthly	River Granta catchment, UK	160	Gomme et al. (1991)
Lindane		1.4–52.2 μg/kg	nonpoint sources	10	single	10 sites, Red River, Vietnam	>5 000	Am et al. (1995)
Lindane		0.1–1 μg/kg	nonpoint sources	7	single	five streams and ditches, southern UK	approx. 10	House et al. (1991)
Lindane		1.1–11.5 μg/kg	nonpoint sources	48	monthly	four sites, northern France	–	Gueune and Winnett (1995)
Lindane		0.2–15.1 μg/kg SP	runoff	18	14 d	Ohebach, northern Germany	1	Liess et al. (1999)
Lindane		3.7–52.8 μg/kg	runoff	3	single	ponds, Ontario, Canada	approx. 5	Bishop et al. (2000)
Malathion	diethyl (dimethoxythiophosphorylthio)succinate	0.1–3 μg/L	application to rice fields	11	14 d	Sawato River, Japan	6	Takamura et al. (1991b)
Malathion		0.26–0.69 μg/L	application to rice fields	3	monthly	Sunagawa River system, Japan	approx. 50	Takamura et al. (1991a)
Methidathion	*S*-2,3-dihydro-5-methoxy-2-oxo-1,3,4-thiadiazol-3-ylmethyl *O,O*-dimethyl phosphorodithioate	0.03–9.2 μg/L	runoff	6	event	San Joaquin tributaries, California	50–100	Domagalski et al. (1997)
Methidathion		0.01–0.6 μg/L	runoff	approx. 60	daily	Sacramento–San Joaquin catchment, California	approx. 40 000	Kuivila and Foe (1995)
Oxamyl	*N,N*-dimethyl-2-methylcarbamoyloxyimino-2-(methylthio)acetamide	0.4–0.6 μg/L	leaching, runoff	3	monthly	seven sites, Pajaro River estuary, California	approx. 140	Hunt et al. (1999)
Oxydemeton-methyl	*S*-2-ethylsulfinylethyl *O,O*-dimethyl phosphorothioate	1–63 μg/L	aerial application	10	event	two vineyard catchments, southwestern Germany	0.1–1.5	Aufseß et al. (1989)
Parathion-ethyl	*O,O*-diethyl *O*-4-nitrophenyl phosphorothioate	0.06–0.4 μg/L	nonpoint sources	4	monthly	six farm ditches, British Columbia, Canada	approx. 0.5	Wan et al. (1994)
Parathion-ethyl		0.3–83 μg/L	aerial application	8	event	two vineyard catchments, southwestern Germany	0.1–1.5	Aufseß et al. (1989)
Parathion-ethyl		3.3–13 μg/kg SP	nonpoint sources	3	single	River Windrush catchment, southern UK	approx. 150	House et al. (1992)
Parathion-ethyl		0.3–1 μg/kg	nonpoint sources	3	single	River Windrush catchment, southern UK	approx. 150	House et al. (1992)
Parathion-ethyl		0.04–6 μg/L	runoff	10	event	Ohebach, northern Germany	1	Liess et al. (1999)
Parathion-ethyl		0.05–50.8 μg/kg SP	runoff	18	14 d	Ohebach, northern Germany	1	Liess et al. (1996)
Parathion-ethyl		1.0–8.7 μg/kg	runoff	3	event	Ohebach, northern Germany	1	Liess et al. (1999)
Parathion-methyl	*O,O*-dimethyl *O*-4-nitrophenyl phosphorothioate	0.001–0.012 μg/L	nonpoint sources	16	60 d	Ioannina Lake, Greece	1 330	Albanis et al. (1986)
Parathion-methyl		0.002–0.032 μg/L	nonpoint sources	9	60 d	Kalamas River, Greece	1 330	Albanis et al. (1986)
Parathion-methyl		0.4–213 μg/L	aerial application	10	event	two vineyard catchments, southwestern Germany	0.1–1.5	Aufseß et al. (1989)
Parathion-methyl		0.01–0.49 μg/L	runoff	7	biweekly	Moon Lake catchment, Mississippi	166	Cooper (1991b)

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Table 1. Continued.

Substance	Chemical name	Concentration†	Source	Number of detections	Sampling interval	Location	Catchment size	Reference
							km²	
Permethrin	3-phenoxybenzyl (1 *RS*,3*RS*;1*RS*,3*SR*)-3-(2,2 dichlorovinyl)-2,2- dimethylcyclo-propanecarboxylate	0.6 μg/L Maximum	nonpoint sources	1	monthly	up to 29 streams, southern Sweden	2–500	Kreuger and Brink (1988)
Permethrin		0.01–0.13 μg/L	runoff	2	biweekly	Moon Lake catchment, Mississippi	166	Cooper (1991b)
Permethrin		0.5–1.6 μg/L (water + particles)	runoff	–	event	South River coastal area, North Carolina	20–50	Kirby-Smith et al. (1992)
Permethrin		2 μg/kg SP	nonpoint sources	1	seasonal	Vemmenhög catchment, southern Sweden	9	Kreuger et al. (1999)
Permethrin		1–3 μg/kg	nonpoint sources	3	seasonal	Vemmenhög catchment, southern Sweden	9	Kreuger et al. (1999)
Permethrin		0.5–163 μg/kg cores	nonpoint sources	5	single	Whitewater River, southern UK	356	Daniels et al. (2000)
Permethrin-trans		18 μg/kg	nonpoint sources	1	single	five streams and ditches, southern UK	approx. 10	House et al. (1991)
Pirimicarb	2-dimethylamino-5,6-dimethylpyrimidin-4-yl dimethylcarbamate	0.48 μg/L	nonpoint sources	1	single	orchard wetlands, Ontario, Canada	approx. 5	Harris et al. (1998)
Pirimicarb		0.1–10 μg/L	nonpoint sources	61	daily	Vemmenhög subcatchment, southern Sweden	8.3	Kreuger (1998)
Pirimicarb		3.7 μg/L (maximum)	nonpoint sources	5	monthly	up to 29 streams, southern Sweden	2–500	Kreuger and Brink (1988)
Pirimicarb		0.2–1 μg/L	runoff	5	daily	Vemmenhög catchment, southern Sweden	9	Kreuger (1995)
Pirmicarb		27.3 μg/L	nonpoint sources	1	event	Riedgraben, southern Germany	6	Schlichtig et al. (2001)
Prothiofos	*O*-2,4-dichlorophenyl *O*-ethyl *S*-propyl phosphorodithioate	0.04–0.16 μg/L	runoff	2	event	Franschoek River, South Africa	20	Schulz (2003)
Prothiofos		15–980 μg/kg SP	runoff	2	event	Lourens River and tributaries, South Africa	92	Schulz (2001b)
Prothiofos		0.5–6 μg/kg SP	runoff	2	14 d	Lourens River tributary, South Africa	0.15	Schulz and Peall (2001)
Pyridafenthion	*O*-(1,6-dihydro-6-oxo-1-phenylpyridazin-3-yl) *O*,*O*-diethyl phosphorothioate	0.1–2 μg/L	nonpoint sources	14	seasonal	Naka River, Japan	991	Kikuchi et al. (1999)
Terbufos	*S*-*tert*-butylthiomethyl *O*,*O*-diethyl phos-phorodithioate	0.013–0.048 μg/L	runoff	–	event	streams in the U.S. Midwest	200–443 670	Battaglin and Fairchild (2002)
Terbufos		0.01–0.3 μg/L	runoff	–	event	Estuarine wetlands, North Carolina	20–50	Kirby-Smith et al. (1992)
Thiobencarb	*S*-4-chlorobenzyl diethylthiocarbamate	0.2–8 μg/L	application to rice fields	4	14 d	Sawato River, Japan	6	Takamura et al. (1991b)
Thiobencarb		2.56 μg/L	application to rice fields	1	monthly	Sunagawa River system, Japan	approx. 50	Takamura et al. (1991a)
Toxaphene	2,2-dimethyl-3-methylennorbornane	2.4–3.9 μg/L	leaching, runoff	2	monthly	seven sites, Pajaro River estuary, California	approx. 140	Hunt et al. (1999)
Toxaphene		0.01–1.2 μg/L	nonpoint sources	60	monthly	five sites, Dear Creek, Mississippi	44	Cooper et al. (1987)
Trichlorfon	dimethyl 2,2,2-trichloro-1-hydroxyethyl-phosphonate	6–182 μg/L	aerial application	18	event	two vineyard catchments, southwestern Germany	0.1–1.5	Aufseß et al. (1989)

† If not mentioned otherwise, the concentrations given refer to the sum of all isomers. SP, suspended particles.

parathion-methyl (0.49 μg/L) originating from cotton (*Gossypium hirsutum* L.), soybean [*Glycine max* (L.) Merr.], and rice (*Oryza sativa* L.) farming, which were found sporadically in water and sediments and in 26% of the fish samples (Cooper, 1991a, 1991b). Along with results from South Carolina estuarine waters (Baughman et al., 1989), these are probably among the first studies from the United States detecting pyrethroids in field samples (Table 1). Much of this work was later reviewed and summarized in various papers (Cooper, 1990; Cooper and Lipe, 1992; Schreiber et al., 1996; Smith et al., 1995). A very early example of the variation of pesticide contents during a spring discharge event was documented for Shell Creek in Nebraska by Spalding and Snow (1989). Based on nine different herbicides and the insecticide disulfoton, this study indicated that the pesticide levels peak before the peak in stream discharge.

Between 1985 and 1987, Kreuger and Brink (1988) conducted a pesticide monitoring program in up to 29 streams with varying catchment sizes in southern Sweden. The organochlorines endosulfan and lindane, the organophosphate fenitrothion, the pyrethroid permethrin, and the carbamate insecticide pirimicarb were detected at maximum levels of 0.1, 0.6, 0.1, 0.6, and 3.7 μg/L, respectively (Table 1). As surface water was estimated to supply 50% of the Swedish drinking water, there was great concern about nonpoint-source pollution of this important resource. Follow-up studies focused on streams and ponds in the Vemmenhög catchment in southern Sweden, which is dominated by winter rape (*Brassica napus* L.), winter wheat (*Triticum aestivum* L.), sugar beet (*Beta vulgaris* L.), and spring barley (*Hordeum vulgare* L.). They detected cyfluthrin, dimethoate, pirimicarb, and permethrin in water samples as well as permethrin and fenvalerate in sediments and suspended particles, respectively (Kreuger, 1995, 1998; Kreuger et al., 1999). These investigations suggested a correlation between amounts used in the catchment and occurrence in water samples and reported a decrease of overall detections between 1990 and 1996. However, concentrations of cyfluthrin, dimethoate, and pirimicarb were transiently above levels demonstrated as having an effect on the aquatic fauna (Kreuger, 1998).

Scott and coworkers conducted extensive field studies in an area of repeated fish kills (Scott et al., 1987; Trim and Marcus, 1990) related to pesticides used on vegetable crops adjacent to estuarine marshes in South Carolina (Scott et al., 1989). An early study linked runoff-related fenvalerate levels up to 0.11 μg/L to in situ toxicity, using shrimp (*Palaemonetes pugio*) (Baughman et al., 1989). Peak field exposures measured between 1985 and 1990 reached 0.85 μg/L for endosulfan, 0.9 μg/L for fenvalerate, and 7 μg/L for azinphos-methyl (Finley et al., 1999; Ross et al., 1996; Scott et al., 1999).

House et al. (1991) were able to detect the pyrethroids cypermethrin, deltamethrin, and permethrin (trans isomer) at levels up to 2.7, 37.5, and 18 μg/kg, respectively, in sediments of ditches, streams, and drainage channels in the southern UK (Table 1). Another study focusing on suspended particles reported considerable levels of dieldrin, DDT, and parathion-ethyl (House et al., 1992), while more recent papers from this group have reported permethrin in sediment cores (Daniels et al., 2000) or concentrated on other insecticide sources, such as the textile industry (House et al., 2000). An extensive study of pesticide transport was conducted at the Agricultural Development and Advisory Service (ADAS) farm at Rosemaund, UK between 1990 and 1992. A total of 59 rainfall–pesticide–location combinations were monitored, during which several herbicides and the insecticide carbofuran were detected at concentrations up to 49.4 μg/L (Table 1; Williams et al., 1995).

In various investigations the rice insecticides carbaryl, diazinon, dimethoate, fenthion, and pyridafenthion were found at levels of <1 μg/L in surface water samples from Japan (Table 1). Additionally, fenobucarb, fenithrothion, fenthion, malathion, and thiobencarb were reported at higher levels of 36.1, 1.7, 50, 3.0, and 8.0 μg/L, respectively (Hatakeyama and Yokoyama, 1997; Iwakuma et al., 1993; Kikuchi et al., 1999; Tada and Hatakeyama, 2000; Tada and Shiraishi, 1994; Takamura, 1996; Takamura et al., 1991b; Tanabe et al., 2001). A small headwater stream situated in a intensively cropped (winter wheat, sugar beet) area in northern Germany was monitored using various sampling techniques to detect insecticides in runoff water, stream water, and suspended particles during runoff events (Liess et al., 1996; Schulz et al., 1998). Transient peak contaminations of fenvalerate (6.2 μg/L and 302 μg/kg in suspended particles) and parathion-ethyl (6.0 μg/L and 50.8 μg/kg in suspended particles) were measured during runoff events between 1992 and 1995 (Liess et al., 1999).

Surface waters in orchard-dominated areas of the Central Valley in California, USA were studied for insecticide input and transport from smaller subcatchments via the San Joaquin and Sacramento River through to the San Francisco Bay. Initial studies focused on diazinon, methidathion, and DDT (Domagalski and Kuivila, 1993; Kuivila and Foe, 1995). Selected storm events monitored by Domagalski et al. (1997) as part of the National Water Quality Assessment program were shown to result in levels of 0.26 μg/L chlorpyrifos, 7 μg/L diazinon, and 9.2 μg/L methidathion in small headwater streams (Table 1). Another study using a surface water monitoring network suggested that the western valley was the principal source of pesticides to the San Joaquin River during the irrigation season (Domagalski, 1997). More recent studies emphasized either the toxicity of insecticide input events (e.g., 4.8 μg/L chlorpyrifos) to water flea (*Ceriodaphnia dubia*) (Amphipoda) (Werner et al., 2000) or the residues of insecticides, such as chlorpyrifos (2.1 μg/kg) and endosulfan (24.6 μg/kg), in suspended particles (Bergamaschi et al., 2001).

Another fruit orchard area has been observed for current-use insecticides since 1998 in the Western Cape of South Africa. High peak concentrations of azinphos-methyl (1.5 μg/L and 1247 μg/kg), chlorpyrifos (0.2 μg/L and 924 μg/kg), endosulfan (2.9 μg/L and 12082 μg/kg), and prothiofos (980 μg/kg) were detected in water and suspended particles of the Lourens River (Table 1) in association with a single storm runoff event during the spraying season (Schulz, 2001b). The first rainfall events of the wet season, occurring about 2 to 3 mo following the last pesticide application, transported mainly particle-associated insecticides (Table 1) via the tributaries into the Lourens River (Dabrowski et al., 2002a; Schulz et al., 2001a). Spray drift was identified as another route of insecticide input, although relatively high levels were detected mainly in the affected tributaries (Schulz et al., 2001b). The monitoring data were recently compared with predictions using basic drift data and a runoff formula suggested by the Organisation for Economic Co-Operation and Development (OECD) (Dabrowski et al., 2002b; Dabrowski and Schulz, 2003). It was demonstrated that runoff is a more important route of pesticide entry than spray drift, producing higher insecticide concentrations and loads in the Lourens River (Dabrowski and Schulz, 2003; Schulz, 2001a).

Some of the insecticides listed more frequently in Table 1 (e.g., chlorpyrifos, azinphos-methyl, diazinon) are among the most heavily used insecticides in the USA (National Center for Food and Agricultural Policy, 1997). However, very little field data exists for aquatic concentrations of other chemicals that are applied in relatively high total amounts in the USA, such as aldicarb, malathion, or carbaryl. However, the mere fact that some chemicals have been studied more frequently than other compounds, or are used intensively in agriculture, by no means justifies a suspicion that these chemicals pose a greater threat to aquatic ecosystems.

In particular, the earlier exposure studies covered in this review did not further specify the routes of nonpoint-source insecticide entry. In total, 27 studies mentioned in Table 1 simply assume agricultural nonpoint sources as the route of entry, of which 20 refer to the period before 1999. Runoff represents by far the most important specified source of insecticide entry, having received increased attention during the past few years as indicated by the high proportion of studies (15 out of 23) published since 2000. Interestingly, only four studies specify spray drift as the route of entry of insecticides (azinphos-methyl, cypermethrin, endosulfan) detected in surface waters, two of them done in the 1980s and the other two in 2001. This lack of field data is surprising in view of the importance of spray drift as an exposure scenario in the regulatory risk assessment scheme of many countries (Aquatic Effects Dialogue Group, 1992; Ganzelmeier et al., 1995; Groenendijk et al., 1994; USEPA, 1999a). However, some studies have addressed the effect of spray depositions due to aerial application of pesticides (Bird et al., 1996; Ernst et al., 1991). Detection of insecticides following application in rice fields was reported in seven studies, two of which appeared since 2000, and leaching was mentioned in two studies from 1998 and 1999.

A few studies reported detections of the same compound in both sediment and suspended-particle samples from the same catchment (Table 1), suggesting higher levels in suspended particles. The chemicals DDT, dieldrin, and parathion-ethyl were detected at levels of 0.2, 0.2, and 1 μg/kg in bottom sediments and at considerably higher levels of 4.7, 17, and 13 μg/kg, respectively, in suspended particles (House et al., 1992). In other studies, fenvalerate and parathion-ethyl reached 71.3 and 8.7 μg/kg in sediments and 302 and 50.8 μg/kg, respectively, in suspended particles (Liess et al., 1996, 1999). However, the distribution of a chemical between sediment and suspended particles is dependent on numerous factors, such as the route of entry into the system and the time between input and sampling. Kreuger et al. (1999) found no clear difference in concentrations between suspended particles and sediments for the pyrethroids permethrin and fenvalerate.

A study undertaken by Kreuger and Brink (1988) on running waters draining catchments of different sizes in a localized area in southern Sweden suggested higher pesticide concentrations in smaller catchments (<100 km^2) than in larger ones. However, this result was mainly derived from herbicide data, as this group of pesticides was most often detected in the various catchments. To analyze for a relationship applicable to insecticides, all insecticide data derived from water samples that are contained in Table 1 were correlated with the average catchment-size information also included in Table 1. The result was that the log-transformed maximum insecticide concentration is negatively correlated (with a significance of $p = 0.0025$) with the log-transformed catchment size (Fig. 1). All 19 detections of a single insecticide concentration of >10 μg/L were obtained in surface water with a catchment size below 100 km^2, indicating the importance of catchment size. Thirteen of these 19 detections of >10 μg/L were obtained in surface water with a catchment size below 10 km^2. This is of particular importance with regard to the European Water Framework Directive (European Union, 2000), which currently only covers >10-km^2 catchments. This important directive thus generally excludes aquatic habitats that are potentially at the highest risk of being negatively affected by high insecticide concentrations.

It is also interesting to note that 12 of these 19 detections of extremely high concentrations (>10 μg/L) resulted from an event-triggered sampling program. This is not surprising; insecticides originating from nonpoint sources are present for only brief periods in small headwater environments and detection would not be possible without using event-controlled sampling (Liess and Schulz, 2000). Insecticide contents in sediments or suspended particles as represented by the studies in Table 1 did not correlate significantly with catchment size, but the data available may not be sufficient to show any reliable trend.

EFFECTS

Table 2 lists the available field studies on biological effects of agricultural insecticide pollution in surface waters. The reports are sorted chronologically to show the historical development. A classification was undertaken based on the relationship between exposure and effects, consisting of the following four classes: "no relation," "assumed relation," "likely relation," and "clear relation." This classification is largely based on the cited authors' judgement of their own results; a relationship

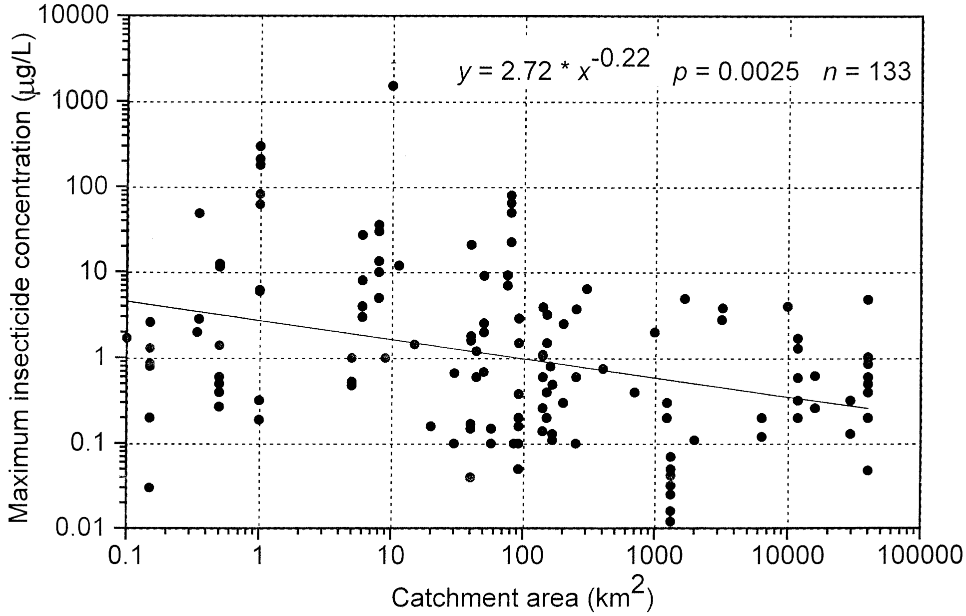

Fig. 1. Relationship between catchment size and aqueous-phase nonpoint-source insecticide contamination detected in samples of surface waters. The database is derived from the studies summarized in Table 1.

was classified as clear only if the exposure was quantified and the effects were linked to exposure temporally and spatially, including control situations without effects. A second important criterion for the evaluation of studies in Table 2 is the exposure scenario. Studies using experimental exposure (insecticide injection or overspraying) were judged less relevant than studies on nonpoint-source pollution events monitored during normal farming practice, and were thus less represented in Table 2. Third, a distinction is made between effects on organisms exposed in situ, which again reflects a more artificial experimental scenario, and effects on parameters such as temporal (abundance) or spatial (drift) species or community dynamics in the field, which are described using the respective sampling methods.

The first studies were reported in the 1970s and 1980s. Many of the early studies were based in Canada and focused on side effects either of aerial application of insecticides (e.g., fenitrothion) to forest environments (Eidt, 1975; Flannagan, 1973; Poirier and Surgeoner, 1988) or of experimental injections of simulium larvicides (e.g., methoxychlor or fenthion) into headwater streams (Burdick et al., 1968; Clark et al., 1987; Cuffney et al., 1984; Dosdall and Lehmkuhl, 1989; Flannagan et al., 1979; Freeden, 1974, 1975; Haufe et al., 1980; Hynes and Wallace, 1975; Wallace and Hynes, 1975; Wallace et al., 1976). They are thus not reported in Table 2. Yasuno et al. (1981) studied the effects of the simulium larvicide temephos, which was experimentally added to two small tributaries of the Yamaguchi River, Japan, on invertebrate drift. Jacobi (1977) developed field containers for the in situ exposure of invertebrates to test for side effects of antimycin applied to kill rough fish. The effects of the lampricide TFM (3-trifluormethyl-4-nitrophenol) on drift and abundance of various insect species were reported by Dermott and Spence (1984).

Cooke (1981) exposed tadpoles of common frogs (*Rana temporaria*) in situ in streams beside potato (*Solanum tuberosum* L.) fields and detected increased rates of deformities after oxamyl application (Table 2). As the exposure concentrations were not measured in this study, it is difficult to establish a link between exposure and effects. Heckman (1981) between 1978 and 1980 performed an extensive survey of the macroinvertebrate fauna in ditches draining an intensively used orchard area in northern Germany and compared his data with results from another study (Garms, 1961) done in the same area between 1951 and 1957, before the commencement of insecticide, acarizide, fungicide, and herbicide application. He concluded that the 25 yr of pesticide application had a major effect in that various insect species, namely 48 of the original 62 coleopteran species (e.g., Dytiscidae and Helodidae), disappeared. On the other hand, Turbellaria were not affected and dipterans even increased in species number. As there were no measurements of insecticide concentrations in water or sediment, a direct cause–effect relationship remains speculative. However, residues of the organochlorine insecticides lindane and DDT have been found in selected invertebrate and fish species (Heckman, 1981), suggesting that aquatic communities have been exposed to these pesticides.

As part of a larger investigation on cypermethrin, Crossland et al. (1982) studied the effects of spray drift–borne residues in a small stream in France during application to adjacent vineyards. Concentrations peaked at 1.7 μg/L and fell to zero within a few hours. Another study examined the effects of aerial application of cypermethrin in drainage ditches bordering winter wheat fields, and found peak levels of 0.03 μg/L (Shires and Bennett, 1985). Both studies concluded that there were no marked biological effects of the transient insecticide contamination on invertebrates, zooplankton, and caged fishes apart from a slight increase in invertebrate drift.

Table 2. Field and in situ studies designed to establish a relationship between the insecticide contamination of surface waters due to usual agricultural practice and effects on the aquatic fauna.

	Insecticide exposure			Toxicological effects		Relationship	
Source	Substance(s)	Quantification	Duration	Endpoint	Species	exposure and effect	Reference
Runoff	oxamyl	no	unclear	deformities (in situ bioassay)	frog (*Rana temporaria*)	assumed	Cooke (1981)
Runoff	unclear	no	unclear	abundance	various invertebrate species	assumed	Heckman (1981)
Spray drift	cypermethrin	0.4–1.7 µg/L	few h	abundance, drift	various invertebrate species	no	Crossland et al. (1982)
Aerial application	cypermethrin	0.03 µg/L	few h	abundance	various planktonic and benthic invertebrates	no	Shires and Bennett (1985)
Runoff	parathion-ethyl	0.3–83 µg/L	few h	abundance	various invertebrate species	assumed	Aufseß et al. (1989)
	parathion-methyl	0.4–213 µg/L					
	trichlorfon	6–182 µg/L					
	oxydemeton-methyl	1–63 µg/L					
Runoff	fenvalerate	0.11 µg/L	few h	mortality (in situ bioassay)	shrimp (*Palaemonetes pugio*)	clear	Baughman et al. (1989)
Runoff	unclear	no	unclear	community composition	various invertebrate species	assumed	Dance and Hynes (1980)
Application to rice fields	fenobucarb	1–4 µg/L	few d	abundance	mayfly (*Baetis thermicus*)	assumed	Takamura et al. (1991b)
Application to rice fields	malathion	0.26–0.69 µg/L	few d	abundance	various odonate species	assumed	Takamura et al. (1991a)
Runoff	unclear	no	unclear	abundance, production	caddisfly (*Hydropsyche* spp.)	assumed	Sallenave and Day (1991)
Runoff	DDT	22–220 µg/kg	unclear	deformities	dipteran (Chironomidae)	assumed	Madden et al. (1992)
Application to rice fields	fenobucarb	22.4 µg/L	few h	abundance	dipteran (*Antocha* spp.)	assumed	Tada and Shiraishi (1994)
Spray drift	pyrethroids	no	unclear	die-off	various fish species	assumed	Sályi and Csaba (1994)
Unclear	unclear	no	unclear	abundance	various invertebrate species	assumed	Lenat and Crawford (1994)
Experimental	parathion-methyl	0.5–5.8 µg/L	few d	mortality (in situ bioassay)	dipteran (*Chaoborus crystallinus*)	clear	Bergema and Rombout (1994)
Experimental	parathion-methyl	0.5–5.8 µg/L	few d	mortality (in situ bioassay)	amphipod (*Gammarus* spp.)	clear	deJong and Bergema (1994)
Application to watercress beds	malathion	no	few h	mortality (in situ bioassay)	amphipod (*Gammarus pulex*)	assumed	Crane et al. (1995b)
Unclear	unclear	no	unclear	mortality (in situ bioassay)	amphipod (*Gammarus pulex*), dipteran (*Chironomus riparius*)	assumed	Crane et al. (1995a)
Unclear	anticholinesterase compound	ND	unclear	die-off	freshwater mussels	assumed	Fleming (1995)
				mussel cholinesterase	mussel (*Elliptio complanata*)		
Runoff	carbofuran	0.05–26.8 µg/L	few h	mortality (in situ bioassay)	amphipod (*Gammarus pulex*)	clear	Matthiessen et al. (1995)
Aerial application	carbaryl	0.1–85.1 µg/L	24 h	drift	various invertebrate species	likely	Beyers et al. (1995)
Aerial application	fenthion	0.5–50 µg/L	few d	mortality (laboratory bioassay)	shrimp (*Paratya compressa improvisa*)	likely	Hatakeyama and Yokoyama (1997)
				field communities	various invertebrate species		
Aerial application	fenobucarb, fenitrothion, fenthion	0.2–1.5 µg/L	few h	life cycle	damselfly (*Calopteryx atrata*)	assumed	Takamura (1996)
Runoff	endosulfan	1.44 µg/L	unclear	die-off	various fish species	likely	Ross et al. (1996)
Leaching (irrigation)	chlorpyrifos	0.12 µg/L	approx. 24 h	brain cholinesterase	carp (*Cyprinus carpio*)	likely	Gruber and Munn (1998)
	azinphos-methyl	0.2 µg/L					
Experimental	deltamethrin	0.46 µg/L	unclear	abundance	various invertebrate species	clear	Lahr (1998)
	diflubenzuron	10 µg/L					
	fenitrothion	80 µg/L					
Aerial application	cypermethrin	2–25 µg/kg	>150 d	abundance, emergence	dipteran (Chironomidae)	clear	Kedwards et al. (1999)
Runoff	unclear	no	unclear	mortality (in situ bioassay)	amphipod (*Hyalella azteca*), dipteran (*Chironomus tentans*)	assumed	Tucker and Burton (1999)

Continued next page.

Table 2. Continued.

	Insecticide exposure			Toxicological effects			
Source	Substance(s)	Quantification	Duration	Endpoint	Species	Relationship exposure and effect	Reference
Runoff	parathion-ethyl	2.3–4.4 μg/kg	few h	muscle cholinesterase	sticklebacks (*Gasterosteus aculeatus*)	clear	Sturm et al. (1999)
Runoff	azinphos-methyl	1.42–21 μg/L	few h	die-off abundance	estuarine fish shrimp (*Palaemonetes pugio*)	likely	Finley et al. (1999)
Runoff	parathion-ethyl	6 μg/L	1 h	mortality (in situ stream) abundance	amphipod (*Gammarus pulex*), caddisfly (*Limnephilus lunatus*)	clear	Liess and Schulz (1999)
Runoff	fenvalerate	0.85–6.2 μg/L	1 h	mortality (in situ bioassay) abundance	amphipod (*Gammarus pulex*), caddisfly (*Limnephilus lunatus*)	clear	Schulz and Liess (1999b)
Runoff	azinphos-methyl endosulfan fenvalerate	0.1–7 μg/L 0.01–0.85 μg/L 0.02–0.9 μg/L	few h	mortality (in situ bioassay)	shrimp (*Palaemonetes pugio*), mummichog (*Fundulus heteroclitus*)	clear	Scott et al. (1999)
Runoff	endosulfan	1.3–10 μg/kg SPMD†	unclear	abundance	Ephemeroptera, trichoptera	clear	Leonard et al. (1999)
Runoff	parathion-ethyl	6 μg/L	1 h	abundance, drift, mortality	Trichoptera, other invertebrates	clear	Schulz and Liess (1999a)
Application to rice fields	lindane, diazinon, cypermethrin-α	no	unclear	community composition	various insect taxa	likely	Suhling et al. (2000)
Runoff	endosulfan	SPMD	unclear	abundance	Ephemeroptera, trichoptera	clear	Leonard et al. (2000)
Rice seed coating	fipronil	9.1 μg/L, 5.5 μg/kg	96 h	mortality (in situ bioassay)	crayfish (*Procambarus* spp.)	clear	Schlenk et al. (2001)
Runoff	azinphos-methyl endosulfan	0.8 μg/L 0.2 μg/L	4 h	mortality (in situ bioassay)	dipteran (*Chironomus* spp.)	clear	Schulz and Peall (2001)
Spray drift	azinphos-methyl	0.87 μg/L	1 h	mortality (in situ bioassay)	dipteran (*Chironomus* spp.)	clear	Schulz et al. (2001c)
Runoff	endosulfan	10–318 μg/kg	few h	abundance, drift	various invertebrate species	clear	Jergentz et al. (2004)
Runoff, spray drift	azinphos-methyl chlorpyrifos	0.82 μg/L 344 μg/kg	1–3 h	community composition	Ephemeroptera, other insects	clear	Schulz et al. (2002)
Experimental	parathion-methyl	1–550 μg/L	few d	abundance	various invertebrate species	clear	Schulz et al. (2003b)
Runoff	chlorpyrifos	1.3 μg/L, 89.4 μg/kg	4 h	mortality (in situ bioassay)	dipteran (*Chironomus* spp.)	clear	Moore et al. (2002)
Runoff	chlorpyrifos endosulfan	300–720 μg/kg 4.6–156 μg/kg	few h	mortality (in situ bioassay)	amphipod (*Paramelita nigroculus*)	clear	Schulz (2003)

† Semipermeable membrane device.

In various trials between 1984 and 1986, Aufseß et al. (1989) measured high concentrations of the organophosphate insecticides parathion-methyl, parathion-ethyl, oxydemeton-methyl, and trichlorfon in streams draining vineyards in southwestern Germany. Changes in the abundance of macroinvertebrates over time were also reported but were not clearly attributable to the timing of pesticide contamination, although 50% of the water samples taken were toxic to waterfleas (*Daphnia magna*). Following runoff events, Baughman et al. (1989) detected fenvalerate at levels of up to 0.1 μg/L in estuarine sites in South Carolina. Shrimp (*Palaemonetes pugio*) exposed in situ showed increased mortality rates in comparison with animals exposed at uncontaminated control sites. Thus, this is probably one of the first studies establishing a link between quantified insecticide exposure due to usual farming practice and biological responses (Table 2); however, it does not deal with the dynamics of in-stream species or communities.

In the early 1990s, further studies on the effects of forest insecticide application in Canada, USA, Japan, and Australia on invertebrate abundance, drift, and emergence were published (Davies and Cook, 1993; Griffith et al., 1996; Hatakeyama et al., 1990; Kreutzweiser and Sibley, 1991; Sibley et al., 1991). A series of studies reported effects of experimental injection of the simulium larvicide methoxychlor into headwaters in Canada on functional community structure, secondary production, and particulate matter export (Lugthart and Wallace, 1992; Lugthart et al., 1990; Wallace et al., 1991a, 1995) and documented the subsequent recolonization patterns (Wallace et al., 1991b). The short-term effects of carbosulfan and permethrin on invertebrate drift in the Black Volta, Ghana were described by Samman et al. (1994). These studies are again not included in Table 2 because of the artificial exposure scenario applied.

Sallenave and Day (1991) documented a factor of five difference in the average yearly secondary production of four coexisting hydropsychid species (Trichoptera) in two tributaries in Ontario differing in the intensity of agricultural land use in their surroundings. Lenat and Crawford (1994) and Dance and Hynes (1980) successfully linked different forms of land use including agriculture with the invertebrate community structure. However, again, no pesticide analyses were performed during these studies. The same applies to die-off events of freshwater fish reported from Hungary with pyrethroids as the potential cause (Sályi and Csaba, 1994). Fleming (1995) investigated a freshwater mussel die-off and measured reduced cholinesterase levels in mussel (*Elliptio complananta*), although no anticholinesterase chemicals were detectable. The lack of exposure data in all these studies makes a direct link of observed effects to contamination impossible.

Fish kills in estuarine waters in South Carolina were assumed to be linked to endosulfan concentrations as high as 1.44 μg/L (Ross et al., 1996). Various Japanese studies from the 1990s examined the potential effects of insecticide use in rice fields on odonata, ephemeroptera, and other insect taxa in the receiving streams (Tada and Shiraishi, 1994; Takamura, 1996; Takamura et al., 1991a, 1991b). However, none of these studies established a clear relationship between exposure and invertebrate dynamics, and thus the authors were only able to assume a link of insecticide pollution to the observed effects (Table 2). Hatakeyama and Yokoyama (1997) later tried to link shrimp mortality in water samples taken during aerial application of fenthion to rice fields in the catchment of the Suna River, Japan to the dynamics of the benthic invertebrate communities. No clear connection was established, since the community structure had already changed between April and May, whereas the first spraying associated with shrimp mortality did not occur until July.

Studies from the UK on in situ exposure of scud (*Gammarus pulex*) (Amphipoda) alone (Crane et al., 1995b) or in combination with the dipteran species midge (*Chironomus riparius*) (Crane et al., 1995a) again only assumed a link to insecticide pollution, as no exposure quantification was conducted. As part of a project on the development of field bioassays in the Netherlands, in situ effects on scud (*Gammarus* spp.) or the dipteran phantom midge (*Chaoborus crystallinus*) (Bergema and Rombout, 1994; deJong and Bergema, 1994) were shown; however, the sites were experimentally polluted. In a stream at the Rosemaund farm in the UK, Matthiessen et al. (1995) observed a high mortality of scud (*G. pulex*) exposed in situ during a runoff-related peak of carbofuran contamination, reaching a level of 264 μg/L. This study is thus only the second example of a clear link between non-experimental, quantified exposure and effects under field conditions, and again employed in situ exposure of the organisms.

Since the late 1990s, only few further studies on the effects of simulium larvicides on aquatic ecosystems have been performed (Crosa et al., 1998). In situ bioassays using an amphipod (*Hyalella azteca*) or midge (*Chironomus tentans*) were developed in the USA and set a precedent for detecting agricultural nonpoint-source pollution in a study without parallel pesticide analyses (Tucker and Burton, 1999). Three insecticides applied to rice fields in southern France were suggested as an important factor determining macroinvertebrate composition (Suhling et al., 2000), although no analytical quantification took place. Finley et al. (1999) reported a fish-kill in South Carolina estuarine waters due to a level of 1.4 μg/L azinphos-methyl. However, even much higher concentrations of this insecticide were measured in the same estuarine waters, and a correlation of the exposure concentrations with reduced shrimp densities and biomass was thus likely (Finley et al., 1999). Lahr (1998) summarized a set of studies undertaken by experimental injection of insecticides into natural ponds as part of a program aimed at assessing the risk assessment of insecticides used in desert locust (*Schistocerca gregaria*) control. The pyrethroid deltamethrin, the organophosphate fenitrothion, and the insect growth regulator diflubenzuron were shown to affect the abundances of various invertebrate species in the ponds. Clear effects on invertebrates were also obtained from a study using wetlands in Mississippi subjected to experimental parathion-methyl contamination as part of a larger investigation on the effects of wetland plants on pesticide transport and toxicity (Schulz et al., 2003b). Clear transient effects on dipterans (chironomids) were observed in a 3.4-ha farm pond experimentally contaminated with spraydrift-borne cypermethrin at levels up to 25 μg/kg in hydrosoils (Kedwards et al., 1999).

As an example using biochemical markers as endpoints, Gruber and Munn (1998) found reduced brain cholinesterase levels in common carp (*Cyprinus carpio*) in a pond in the central Columbia Plateau, USA that had been affected by organophosphates presumably introduced via leaching as a result of irrigation. As the measured concentrations were $\leq$0.2 μg/L, the authors did not establish a direct link between exposure and effects. In contrast, a study on cholinesterase activities in three-spined sticklebacks (*Gasterosteus aculeatus*) showed a clear link to measured parathion-ethyl concentrations between 2.3 and 4.4 μg/L (Sturm et al., 1999). Of the nine headwater streams studied in northern Germany during this investigation, only two were contaminated with parathion-ethyl.

Several studies undertaken during the past five years have successfully linked survival of in situ exposed organisms to quantified insecticide contamination. Scott et al. (1999) employed bioassays with shrimp (*P. pugio*) and mummichog (*Fundulus heteroclitus*) to detect effects of transient contamination by azinphos-methyl, endosulfan, and fenvalerate introduced into South Carolina estuarine waters via runoff. Kirby-Smith et al. (1989) found no effects in field-deployed shrimp (*P. pugio*) at concentrations below the laboratory effects levels. Chironomids and an indigenous amphipod species (*Paramelita nigroculus*) were established as in situ exposure bioassays for the assessment of aqueous-phase and particle-associated insecticide (azinphos-methyl, chlorpyrifos, endosulfan) toxicity in orchard rivers in the Western Cape of South Africa (Moore et al., 2002; Schulz and Peall, 2001; Schulz et al., 2001c; Schulz, 2003). The response of crayfish (*Procambarus* spp.) exposed in situ to fipronil used as a rice seed coating in Mississippi is reported by Schlenk et al. (2001). The validity and ecological relevance of an in situ bioassay was tested by Schulz and Liess (1999b) in an agricultural headwater stream in northern Germany during runoff-related contamination with fenvalerate up to 6.2 μg/L. Caddisfly (*Limnephilus lunatus*) and amphipod (*G. pulex*) both showed mortality in the in situ bioassays during transient insecticide pollution. However, the authors inferred from their results that in situ bioassays using mobile species such as amphipods may overestimate field toxicity, as the caged organisms are prevented from performing any avoidance reactions, such as downstream drift. Another study in the same catchment used a set of microcosms fed by stream water, of which some were run in a closed circuit during runoff-related parathion-ethyl and fenvalerate exposure in the stream, to show effects on the same two invertebrate species (Liess and Schulz, 1999). Both studies successfully linked their experimental results to the abundance dynamics of the same species in the stream itself. Similarly, a link between survival in multispecies microcosm exposed to azinphos-metyl and the abundance dynamics of invertebrate species at various sites in a transiently insecticide-contaminated river system was recently established in the Lourens River catchment, South Africa (Schulz et al., 2002).

Leonard et al. (1999) studied the abundance of six invertebrate species at eight sites in the Namoi River, southeastern Australia in relation to the cotton insecticide endosulfan, which enters the water mainly via runoff. This data set was extended in a second study and analyzed with different multivariate statistical procedures including principal response curves (Leonard et al., 2000). The results of both studies indicate links between the dynamics of the six dominant species and the endosulfan contamination; however, the pesticide contamination was measured using solvent-filled polyethylene bags as passive samplers and endosulfan was not quantified in water samples directly. The endosulfan concentrations in the passive samplers were correlated with endosulfan levels in bottom sediments, indicating field concentrations up to 10 μg/kg in the sediments (Leonard et al., 1999). Rather high levels of endosulfan between 10 and 318 μg/kg detected in suspended particles in rural rivers near Buenos Aires, Argentina were recently shown to affect the abundance dynamics and drift of various insect species (Jergentz et al., 2004). During this study, two contaminated rivers showed decreased abundances of mayfly and dragonfly species along with drift peaks, while a third river served as an uncontaminated control with unaffected population dynamics. Three sites in a headwater stream in northern Germany were used to measure the abundance and drift of macroinvertebrates (Schulz and Liess, 1999a). Out of the total of eleven core species, eight disappeared following a runoff-related peak concentration of 6 μg/L parathion-ethyl in water samples. A large increase in drift and an elevated mortality rate for caddisfly species in the drift added further evidence indicating the insecticide exposure as the responsible factor. Furthermore, the authors were able to show that even stronger rainfall-related runoff events without pesticide contamination that occurred shortly before the insecticide application period had no effects on the invertebrate abundances or drift, suggesting that other parameters such as hydraulic stress or turbidity were of minor importance during this study (Schulz and Liess, 1999a).

It thus follows from Table 2 that since 1999, a total of eight published studies have shown a more or less clear link between agricultural insecticide pollution and abundance dynamics or community composition of macroinvertebrates. Evidently, increased interest in the topic, in combination with the development of more sophisticated methods for sampling and data analysis, have been responsible for the abundance of recent papers successfully linking agricultural insecticide contamination with observed biological effects at the population or community level. However, it is important to note that for almost all of these studies that seem to establish a clear link between exposure and effect, the pesticide concentrations measured in the field were not high enough to support an explanation of the observed effects simply based on acute toxicity data. Matthiessen et al. (1995) observed 100% mortality of caged scud (*G. pulex*) following exposure to a peak concentration of 27 μg/L carbofuran, which exceeded the 24-h LC_{50} of 21 μg/L for only 3 to 5 h. Baughman et al. (1989) suggested differences in measured and real exposure concentrations to be a reason for higher mortalities in in situ bioassays than predicted from laboratory data. The measured short-term peak concentrations of 6 μg/L parathion-ethyl or 6.2 μg/L fenvalerate associated with field effects (Liess and Schulz, 1999; Schulz and Liess, 1999a, 1999b) are also well below laboratory-derived 24-h LC_{50} values with an initial 1-h exposure period (Liess, 1994). Furthermore, although it was suggested that the endosulfan levels up to 10 μg/kg in the Namoi River have deleterious effects on mayfly (*Jappa kutera*) and other invertebrates (Leonard et al., 1999, 2000), these and even the overall peak concentrations of 48 μg/kg obtained from another study are lower than the 10-d LC_{50} of 162 μg/kg for mayfly (Leonard et al., 2001). On the basis of present knowledge, it cannot be determined whether the measured concentrations in the field regularly underestimate the real exposure or if a general difference between the field and laboratory reactions of aquatic invertebrates is responsible for this situation.

Apart from some studies that used experimental pesticide injection, all field studies on insecticide effects listed in Table 2 were undertaken in surface waters that have been receiving insecticide pollution for as long as several years up to a few decades. In ecological science, Connell (1980) once coined the expression "ghost of competition past" with reference to the hypothesis that competition is not recently visible in communities because it has acted in the past in a way that eliminated competition as a driving force for community structure. Accordingly, a "ghost of disturbance past" might cause difficulty in detecting pesticide-related effects in communities recently inhabiting agricultural surface waters, since any potential pesticide influence would have already acted several years ago.

COMBINATION OF EXPOSURE AND EFFECTS

Exposure concentrations detected in the field are commonly compared with surface water quality guidelines for the protection of aquatic life. Table 3 lists existing water quality guidelines for insecticides relevant for short-term or single exposure in comparison with detected concentrations in surface waters according to the studies summarized in Table 1. It becomes evident that for all insecticides listed, apart from cypermethrin, cyfluthrin, dieldrin, thiobencarb, and terbufos, the established water quality guideline has been exceeded in multiple instances. The exceeding factor lies between 3 and 5 orders of magnitude for most of the insecticides. Azinphos-methyl, chlorpyrifos, diazinon, and endosulfan were shown to exceed the water quality guideline in 9 to 14 studies extracted from Table 1. A total of at least 88 incidents of contamination above the water quality guidelines were reported. It is likely that even more detections of high peak insecticide levels would have been reported, if more than only 50% of the studies listed in Table 1 had employed an event-triggered sampling design.

Table 3. Water quality guidelines for selected insecticides in comparison with maximum concentrations found in surface waters and number of studies reporting exceedence of the respective water quality guideline.

Substance	Water quality guideline†	Maximum detected concentration‡	Number of studies exceeding quality guideline‡
	μg/L		
Aldicarb	1	6.4	2
Azinphos-methyl	0.01	21	9
Carbaryl	0.02	85.1	3
Carbofuran	0.5	49.4	3
Chlorpyrifos	0.01	3.8	11
Cypermethrin	0.0068	1.7	1
Cyfluthrin	0.0015	5.0	1
DDT	0.01	1.1	2
Deltamethrin	0.0004	2	2
Diazinon	0.01	7	11
Dieldrin	0.24	0.26	1
Dimethoate	6.2	30	2
Endosulfan	0.01	1530	14
Fenitrothion	0.11	1.7	3
Fenvalerate	0.008	6.2	4
Lindane	0.01	4.9	4
Malathion	0.1	3	2
Parathion-ethyl	0.011	83	3
Parathion-methyl	0.012	213	3
Permethrin	0.0065	1.6	3
Terbufos	0.003	0.3	1
Thiobencarb	3.1	8.0	1
Toxaphene	0.0002	3.9	2

† Short-term or single application values derived from Brock et al. (2000), Canadian Council of Ministers of the Environment (2001), USEPA (1999b), and California Environmental Protection Agency (2000).

‡ Data are extracted from Table 1.

Table 4 compares the insecticide concentrations having clear effects in field studies as listed in Table 2 with the maximum detected concentrations from exposure field studies in Table 1. Where available, the three highest exposure concentrations measured are included. Each of the values given for a particular insecticide as a measure of effects or exposure is derived from a different study. It is evident that for azinphos-methyl, chlorpyrifos, deltamethrin, endosulfan, and parathion-ethyl the concentrations detected are well above the minima for causing effects in organisms exposed in situ or populations and communities studied in the field. For carbofuran, only one documented exposure value exceeds the reported effect level. For fenvalerate, one exposure concentration exceeds the effect level from one of the effects studies available. No assessment is possible for fipronil or diflubenzuron as the only exposure concentrations available are from the same study that reported the effects, or there are no exposure data.

In summary, this comparison based exclusively on field studies for both exposure and effects suggests that for various insecticides, there exists a potential for effects on the aquatic fauna under natural conditions. The absolute number of 27 potentially critical situations derived from this comparison and the number of nine insecticides for which such a comparison is possible

Table 4. Comparison of insecticide concentrations demonstrated to have clear effects in the field with the three highest concentrations detected for the same insecticide in field studies.†

	Field effect concentrations‡		Detected field concentrations§	
Insecticide	In situ bioassays	Abundance or drift reaction	Water	Sediments or suspended particles
Azinphos-methyl	0.2 μg/L	0.82 μg/L	21 μg/L	
	0.7 μg/L		1.5 μg/L	
			1 μg/L	
Carbofuran	26.8 μg/L		49.4 μg/L	
			4.8 μg/L	
			1.8 μg/L	
Chlorpyrifos	1.3 μg/L	344 μg/kg	3.8 μg/L	924 μg/kg
	300–720 μg/kg		3.2 μg/L	720 μg/kg
	9.4 μg/kg		2.8 μg/L	344 μg/kg
Deltamethrin		0.46 μg/L	2.0 μg/L	
			1.4 μg/L	
			–	
Diflubenzuron	10 μg/L		–	
Endosulfan	0.8 μg/L	318 μg/kg	1530 μg/L	12 082 μg/kg
	4.8–156 μg/kg		13 μg/L	2 461 μg/kg
	10 μg/kg		4 μg/L	318 μg/kg
Fenitrothion	80 μg/L		1.7 μg/L	
			0.4 μg/L	
			0.2 μg/L	
Fipronil	9.1 μg/L		9.1 μg/L	5.5 μg/kg
	5.5 μg/kg			
Fenvalerate	6.2 μg/L	6.2 μg/L	6.2 μg/L	
	0.11 μg/L		0.11 μg/L	
			0.1 μg/L	
Parathion-ethyl	6 μg/L¶	6 μg/L	83 μg/L	50.8 μg/kg
	0.5–5.8 μg/L		6 μg/L	13 μg/kg
	2.3–4.4 μg/kg#		0.4 μg/L	8.7 μg/kg

† Each value given for a particular insecticide for the effects or exposure is derived from a different study.
‡ Data are derived from Table 2.
§ Information on detected concentrations is included for both water and particles only if effective concentrations are also available for both matrices. Data are derived from Table 1. Up to three highest available concentrations are given.
¶ Field stream microcosm result.
# Effect on fish cholinesterase.

might still appear to be quite small. However, it is important to note that the field effect studies that are used in Table 4 are examples that were actually present under normal farming practice. As discussed in the previous section, relatively few and mostly recent studies exist on the effects under field conditions. It is thus possible that further field investigations might reveal further evidence of effects, lower effect levels, or provide results for other insecticides.

RISK MITIGATION

The terms "risk mitigation" and "best management practice" for pesticides are used in a similar way, as general designations of a process in which manufacturers, farmers, and regulators negotiate various sorts of restrictions or alterations of agricultural practice to avoid predicted unacceptable risk. Practical methods of controlling pollution risk have been reviewed, including both in-field (soil conservation measures, application practices, and integrated pest management [IPM]) and end-of-field (buffer zones) techniques (Mainstone and Schofield, 1996). The specific types of pesticide-related best management practices (BMPs) commonly used in the United States include reducing pesticide use, improving the timing and efficiency of application, preventing backflow of pesticides into water supplies, improved calibration of pesticide spray equipment, and IPM (Caruso, 2000).

The effects of conservation tillage are summarized by Fawcett et al. (1994), focusing mainly on herbicides. The Brimstone farm experiment in the UK is described by Harris et al. (1995) as a practical example for the positive effect of agricultural management on pesticide runoff. Programs aiming at changes in the application practices, namely reduced pesticide use, were successfully implemented in Ontario, Canada (Gallivan et al., 2001) and in Norway (Epstein et al., 2001). Measures to reduce pesticide loss due to spray drift have been discussed in relation to IPM by Matthews (1994), while Blommers (1994) summarizes IPM options for apple (*Malus domestica* Borkh.) orchards in Europe. Integrated pest management in general was subjected to a recent review by Way and van Emden (2000). A special program of integrated crop management from the UK covers not only crop protection, but also landscape features, management of the soil, wildlife, and habitats (Drummond and Lawton, 1995). A five-year study by Kirby-Smith et al. (1992) of pesticide runoff and associated ecological effects in an estuarine watershed in North Carolina demonstrated how conservative pest management practices that minimized pesticide application frequency and rates coupled with the use of less persistent pesticides can reduce the toxicity to single species monitored in the field and laboratory tests, and communities of benthic and pelagic invertebrate and fish. Economic aspects of nonpoint-source pollution control measures for the management of environmental contamination by agricultural pesticides have also been summarized (Falconer, 1998; Mainstone and Schofield, 1996).

Buffer zones, in terms of no-spray field margins or noncrop, vegetated riparian strips to prevent pesticide movement from application areas to adjacent nontarget aquatic habitats, have received increasing attention as an agricultural end-of-field best management practice. Based on permethrin applications in Canadian forests, a technique for estimating the width of buffer zone areas during pesticide application based on experimental spray drift data and laboratory toxicity results has been suggested (Payne et al., 1988). Attempts were made in the United States to link knowledge obtained from spray drift studies to buffer width definitions (i.e., to base no-spray zones on spray quality, release height, and other variables, such as wind speed, for protecting specific sensitive areas) (Hewitt, 2000). In 1999, the Local Environmental Risk Assessments for Pesticides (LERAPs) were implemented in the UK, considering the use of reduced application rates, engineering controls, or the size of the watercourse as three factors that might allow some reduction in the no-application zone to be achieved (Ministry of Agriculture, Fisheries and Food, 1999). A recent review on the use of windbreaks as a pesticide drift mitigation strategy concluded that there are still enormous data gaps to be filled before this method can be used efficiently (Ucar and Hall, 2001). Quite apart from water quality considerations, a compelling argument can be made for the establishment of

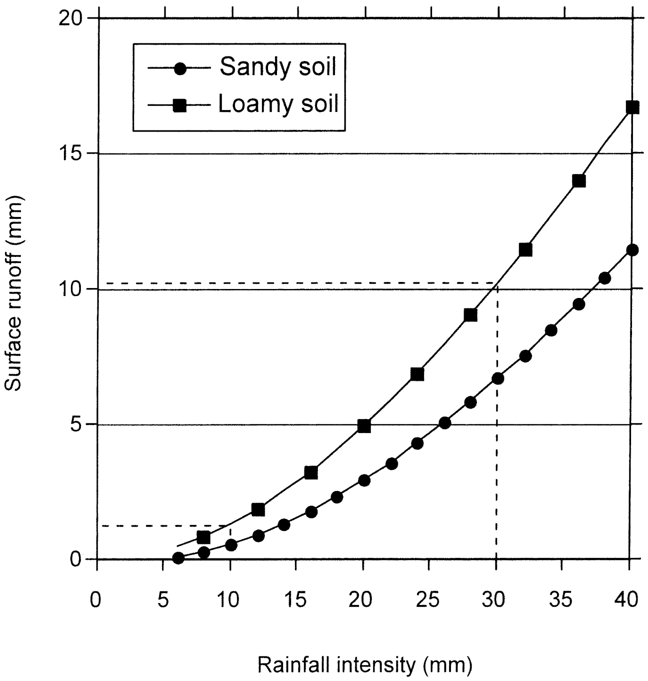

Fig. 2. The amount of surface runoff on sandy and loamy soils with a high soil moisture in relation to rainfall intensity, according to Lutz (1984) and Maniak (1992).

buffer zones in many areas on the basis of their potential for enhancing the ecological quality of river corridors, through the extension of management (e.g., no-spray zones) alongside riverbanks (De Snoo, 1999; Schultz et al., 1995). It is important, however, to recognize that buffer zones are not a solution to the root cause of agricultural contamination of receiving waters, which is related to certain in-field agricultural practices that produce both contaminated runoff and unnecessary aerial transport of contaminants (Mainstone and Schofield, 1996).

As spray deposition decreases exponentially with increasing distance from the sprayed area (Ganzelmeier et al., 1995; USEPA, 1999a), a positive effect of buffer zones on the reduction of drift access to adjacent water bodies and thus the risk to aquatic organisms is very likely and has been shown in field trials (De Snoo and De Wit, 1998). Vegetated buffer strips were also mentioned as a means of reducing runoff-related pesticide transport to surface waters. Auerswald and Haider (1992) investigated copper-containing chemical loss from hops and showed that small particles, which may be associated with a large proportion of pesticide loss during small-sized erosion events (Ghadiri and Rose, 1991), are retained in grassed buffer strips only if they are at least 30 m wide. Experiments conducted in France with different herbicides indicated a reduction in runoff volume by 43 to 99.9% in the presence of grassed buffers strips with widths between 6 and 18 m (Patty et al., 1995, 1997). In a wet 15-m-wide buffer strip, the herbicides isoproturon and pendimethalin were retained by 75 and 96%, respectively (Spatz et al., 1997). However, there are few studies on the retention capabilities of buffer zones for insecticides. In summary, the available results on the effectiveness of buffer zones show buffering capacity for individual contaminants to be variable, largely reflecting the diversity of conditions in which they operate. In addition, the effectiveness of removal under fixed conditions varies depending on chemical characteristics (Mainstone and Schofield, 1996).

According to some authors, the suitability of buffer strips to retain mobile pesticides is questionable (Williams and Nicks, 1993). One aspect that might restrict the effectiveness of any buffer strip is the rather simple relation between rainfall intensity and the amount of water leaving the fields via surface runoff. To illustrate this "hydrological dilemma," the amount of surface runoff was related to rainfall intensity in a simple model on the basis of theoretical considerations and a large amount of empirical data from Germany (Lutz, 1984; Maniak, 1992), as outlined in Fig. 2.

It is evident that an increase in rainfall intensity on loamy soil with a high soil moisture by a factor of three, from 10 to 30 mm, results in an increase of surface runoff by a factor of ten, from about 1 to 10 mm. That means that heavy rainfall events causing storm runoff are always associated with the production of extremely large volumes of water in a short time. In many circumstances these large water volumes may not be retained by any sort of widely employed buffer strip, and erosion channels formed during these conditions may further jeopardize the positive effect of buffer zones. This "hydrological dilemma" may result in unavoidable pesticide contaminations of surface waters specifically under conditions where other measures are not applicable or do not produce the necessary benefit (i.e., high-quality soil areas under intensive agricultural use). In these cases, structural features of the receiving surface waters, such as vegetation coverage, may be useful in mitigating the risk of insecticide pollution.

Constructed wetlands or vegetated ditches have been proposed in this context as risk mitigation techniques. Complementing their ecological importance as ecotones between land and water (Mitsch and Gosselink, 1993) and as habitats with great diversity and heterogeneity (Wetzel, 1993), specifically constructed wetlands are used extensively for water quality improvement. The concept of vegetation as a tool for contaminant mitigation (phytoremediation) is not new (Dietz and Schnoor, 2001). Many studies have evaluated the use of wetland plants to mitigate pollutants such as road runoff, metals, dairy wastes, and even municipal wastes (Brix, 1994; Cooper et al., 1995; Gray et al., 1990; Kadlec and Knight, 1996; Meulemann et al., 1990; Osterkamp et al., 1999; Scholes et al., 1998; Vymazal, 1990). According to Luckeydoo et al. (2002), the vital role of vegetation in processing water passing through wetlands is accomplished through biomass nutrient storage and sedimentation, and by providing unique microhabitats for beneficial microorganisms. Macrophytes serve as filters by allowing contaminants to flow into plants and stems, which are then sorbed to macrophyte biofilms (Headley et al., 1998; Kadlec and Knight, 1996). According to Zablotowicz and Hoagland (1999), whether or not plants are capable of transferring contaminants from environmental matrices depends upon several factors including contaminant chemistry, plant tolerance to the contaminant, and sediment surrounding the plant (e.g., pH, redox, clay content).

Initially wetlands were employed mainly to treat point-source wastewater (Vymazal, 1990), followed later by an increased emphasis on nonpoint-source urban (Shutes et al., 1997) and agricultural runoff (Cole, 1998; Higgins et al., 1993; Rodgers et al., 1999). While the fate and retention of nutrients and sediments in wetlands are understood quite well (Brix, 1994), the same cannot be claimed for agrochemicals (Baker, 1993). Most of the initial studies referred to the potential of wetlands for removal of herbicides and some other organic chemicals (Kadlec and Hey, 1994; Lewis et al., 1999; Moore et al., 2000; Wolverton and Harrison, 1975; Wolverton and McKown, 1976). Since wetlands have the ability to retain and process transported material, it seems reasonable that constructed wetlands, acting as buffer strips between agricultural areas and receiving surface waters, could mitigate the effect of pesticides in agricultural runoff (Rodgers et al., 1999). The effectiveness of wetlands for reduction of hydrophobic chemicals (e.g., most insecticides) should be as high as for suspended particles and particle-associated phosphorus (Brix, 1994; Kadlec and Knight, 1996), since these chemicals enter aquatic ecosystems mainly in particle-associated form following surface runoff (Ghadiri and Rose, 1991; Wauchope, 1978).

Table 5 summarizes the few studies undertaken so far on insecticide retention in constructed wetlands and vegetated ditches. The initial studies attempted to quantify insecticide retention in wetlands by taking input and output measurements and were done on various current-use insecticides in South Africa. Schulz and Peall (2001) investigated the retention of azinphos-methyl, chlorpyrifos, and endosulfan introduced during a single runoff event from fruit orchards into a 0.44-ha wetland. They found retention rates between 77 and 99% for aqueous-phase insecticide concentrations and >90% for aqueous-phase insecticide load between the inlet and outlet of the wetland. Particle-associated insecticide load was retained in the same wetland at almost 100% for all the studied organophosphate insecticides and endosulfan. A toxicity reduction was also documented by midge (*Chironomus* spp.) exposed in situ at the inlet and outlet of the constructed wetland (Table 5). Another study performed in the same wetland assessed spray drift–borne contamination of the most commonly used insecticide, azinphos-methyl, and found similar retention rates; however, the retention rate for the pesticide load was only 54.1% (Schulz et al., 2001c). In parallel, Moore et al. (2001) conducted research on the fate of lambda-cyhalothrin experimentally introduced into slow-flowing vegetated ditches in Mississippi. They reported a more than 99% reduction of pyrethroid concentrations below target water quality levels within a 50-m stretch due to an 87% sorption to plants. A further study demonstrated retention of approximately 55 and 25% of chlorpyrifos by sediments and plants, respectively, in wetland mesocosms (59–73 m in length) in Oxford, Mississippi as well as a >90% reduction in concentrations and in situ toxicity of chlorpyrifos in the wetland in South Africa (Moore et al., 2002).

Another experiment in Oxford, MS targeted the effects of vegetated (>90% macrophyte coverage) versus nonvegetated (<5% macrophyte coverage) wetland mesocosms on the transport and toxicity of parathion-methyl introduced to simulate a worst-case storm event (Schulz et al., 2003b). Both wetland invertebrate communities and midge (*C. tentans*) exposed in situ were significantly less affected in the vegetated wetlands (Table 5) confirming the importance of macrophytes in toxicity reduction. Initial parathion-methyl concentrations of more than 400 μg/L were reduced to below detection limit (0.1 μg/L) within 40 m from the inlet in the vegetated wetlands, while concentrations as high as 8 μg/L were present at 40 m in the nonvegetated wetlands. A parallel study using laboratory testing with amphipod (*Hyalella azteca*) indicated that 44 m of vegetated and 111 m of nonvegetated wetland would reduce the mortality to <5% (Schulz et al., 2003c). The processes relevant for aqueous-phase pesticide dissipation of azinphos-methyl were the subject of another recent study using the flow-through wetland along one of the tributaries of the Lourens River in South Africa (Schulz et al., 2003a). The living plant biomass accounted for 10.5% of the azinphos-methyl mass initially retained in the wetland, indicating processes such as volatilization, photolysis, hydrolysis, or metabolic degradation as being very important.

Apart from these more focused studies, a few further studies are included in Table 5. The implementation of retention ponds in agricultural watersheds was examined by Scott et al. (1999) as one strategy to reduce the amount and toxicity of runoff-related insecticide pollution discharging into estuaries. However, wetland sizes and retention rates are not further detailed. Briggs et al. (1998) inferred, from a study in which nursery runoff was experimentally added to clay–gravel or grass beds of up to 91 m in length, a reduction of >99.9% in terms of the applied chlorpyrifos load, which was not further quantified. A positive effect of settling ponds, situated below watercress (*Nasturtium officinale* R. Br.) beds in the UK that were not further described, was documented using mortality and acetylcholinesterase inhibition in scud (*G. pulex*) exposed in situ as endpoints (Crane et al., 1995b). Retention rates are not given, as the concentrations of malathion used in the watercress beds were not measured in this study.

In summary, very few and only recent studies have dealt with wetlands or vegetated ditches as risk mitigation tools for nonpoint-source insecticide pollution. However, the results obtained thus far on chemical retention and toxicity reductions are very promising (Table 5), and justify further investigation. A few other studies that have emphasized special aspects of pesticide fate or toxicity in wetlands (Dieter et al., 1996; Spongberg and Martin-Hayden, 1997) or uptake of insecticides to plants (Hand et al., 2001; Karen et al., 1998; Weinberger et al., 1982) corroborate the idea that aquatic macrophytes are important to insecticide risk reduction.

Certain agricultural sectors, such as the greenhouse

Table 5. Field studies on the effectiveness of constructed wetlands or vegetated ditches in mitigating agricultural insecticide contamination in surface waters.

Source	Substance	Inlet concentration	Retention: Concentration	Retention: Load	Location	Wetland size	Dominant plant species	Ecotoxicological assessment	Reference
			%	%		m			
Application to watercress beds	malathion	–	–	–	settling ponds below treated watercress beds, UK	–	–	mortality reduction, scud (*Gammarus pulex*) in situ bioassay	Crane et al. (1995b)
Experimental nursery runoff	chlorpyrifos	no data	no data	>99.9†	clay–gravel or grass beds below nursery, South Carolina	2 × 91	bermudagrass [*Cynodon dactylon* (L.) Pers.]	no data	Briggs et al. (1998)
Experimental runoff	lambda-cyhalothrin	500 μg/L	>99	>99	vegetated ditches, Mississippi	50 × 1.5	water persicaria (*Polygonum amphibium* L.), rice cutgrass [*Leersia oryzoides* (L.) Sw.], *Sporobolus* spp.	no data	Moore et al. (2001)
Experimental runoff	chlorpyrifos	73–733 μg/L	no data	83–98	wetland mesocosms, Mississippi	66 × 10	soft rush (*Juncus effusus* L.), *Leersia* spp.	no data	Moore et al. (2002)
Experimental runoff	parathion-methyl	4–420 μg/L	>99	>99	wetland mesocosms, Mississippi	50 × 5.5	soft rush, *Leersia* spp.	>90% toxicity reduction, midge (*Chironomus* spp.) in situ bioassay, reduced effects on invertebrates	Schulz et al. (2003b)
Experimental runoff	parathion-methyl	4–420 μg/L	>99	>99	wetland mesocosms, Mississippi	50 × 5.5	soft rush, *Leersia* spp.	>95% toxicity reduction in laboratory exposed amphipod (*Hyalella azteca*)	Schulz et al. (2003c)
Runoff	azinphos-methyl endosulfan chlorpyrifos azinphos-methyl endosulfan prothiofos	0.14–0.8 μg/L 0.07–0.2 μg/L 0.01–0.03 μg/L 1.2–43.3 μg/kg 0.2–31.4 μg/kg 0.8–6 μg/kg	77–93 >99 >99 >99 >99 >99	>90 >90 >99 >99 >99 >90	flow-through wetland, Lourens River catchment, South Africa	134 × 36	bulrush (*Typha capensis* Rohrb.), shore rush (*Juncus kraussii* Hochst)	>90% toxicity reduction *Chironomus* spp. in situ bioassay	Schulz and Peall (2001)
Runoff	azinphos-methyl endosulfan fenvalerate	0.2–3.9 μg/L 0.03–0.25 μg/L 0.05–0.9 μg/L	>99‡ ‡>60‡ >80‡	no data	retention ponds, South Carolina	no data	no data	approx. 40% toxicity reduction, shrimp (*Palaemonetes pugio*) in situ bioassay	Scott et al. (1999)
Runoff	chlorpyrifos	0.08–1.3 μg/L 2.6–89.4 μg/kg	>97 >99	>97 >99	flow-through wetland, Lourens River, South Africa	134 × 36	bulrush, shore rush	>90% toxicity reduction *Chironomus* spp. in situ bioassay	Moore et al. (2002)
Spray drift	azinphos-methyl	0.27–0.51 μg/L	90.1	60.5	flow-through wetland, Lourens River, South Africa	134 × 36	bulrush, shore rush	reduced effects on zooplankton	Schulz et al. (2003a)
Spray drift	azinphos-methyl	0.36–0.87 μg/L	90.8	54.1	flow-through wetland, Lourens River, South Africa	134 × 36	bulrush, shore rush	>90% toxicity reduction *Chironomus* spp. in situ bioassay	Schulz et al. (2001c)

† Refers to the applied amount.
‡ Estimated retention since the concentrations refer to a catchment without ponds, which was used for comparison.

and nursery industry, have already started to adopt wetlands to treat pesticide-contaminated water (Berghage et al., 1999). In response to the historic losses of natural wetlands, the USDA Natural Resources Conservation Service has established a conservation practice standard (Code 656) relating to constructed wetlands and three standards (Codes 657, 658, and 659) relating to the restoration, creation, and enhancement of natural wetlands (USDA Natural Resources Conservation Service, 2002). By establishing these practice standards, farmers and other agricultural landowners are given instructions on how to develop and use natural and constructed wetlands as a best management practice to minimize nonpoint-source pollution of water bodies.

CONCLUSIONS AND FUTURE RESEARCH DIRECTIONS

On the basis of the literature review presented here, the following conclusions can be drawn and recommendations made with respect to future research.

- Most of the exposure studies refer to runoff as a route of entry. There is a lack of data with respect to surface water contamination by insecticides due to spray drift, which is of particular importance with regard to the role of spray drift in regulatory risk assessment.
- The exposure data emphasizes the high risk to surface waters in small catchments. As a result, future monitoring programs should include catchments of small size, specifically with regard to recent legislative concepts such as the European Water Framework Directive and the U.S. total maximum daily load (TMDL) concept.
- More field studies using event-triggered sampling design are necessary to provide a realistic picture of insecticide levels resulting from nonpoint-sources.
- Most of the insecticide records were made in running water ecosystems, indicating their importance as receiving habitats. One potential reason might be the relatively higher bank length for a river in comparison with the water volume; however, these aspects still need to be addressed. There remains the question whether stagnant water bodies, which play a major role in regulatory risk assessment, run the same risk of exposure under field conditions as streams or rivers.
- The low number of field studies that linked quantified exposure to effects is remarkable. Whatever conclusions may be drawn from this fact in terms of regulatory risk assessment, the lack of field data means that there is also very little understanding of how whole surface water ecosystems react to chemical input.
- For many insecticides that were detected frequently in field samples, knowledge of effects derived from field studies is limited or depends only on results that are not easy to interpret.
- Generally, exposure and effect estimates in ecological risk assessment procedures should be compared with data from field studies under normal agricultural practice, if available, since effects are not always interpretable from laboratory results.
- More wetland research is necessary to increase our understanding of the relevant chemical and biological processes and the long-term sustainability of these systems. Additionally, quantitative results (e.g., on necessary wetland length or effective plant species) are needed to formulate guidelines for the construction and management of these wetlands.
- The definition and implementation of additional risk mitigation strategies and improved measures of their mitigation capabilities might make it possible to adapt the farming and pesticide application practice on a local level (e.g., to reduce or differentiate the distances between sprayed fields and surface waters for specified compounds).

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